Extended Abstract

Exhalation Rate Study of Thoron in Some Building Materials of the Iberian Peninsula †

Samuel Frutos-Puerto 1,*, Eduardo Pinilla-Gil 1, Conrado Miró 2, Eva Andrade 3,4, Mario Reis 3,4 and María José Madruga 3,4

1 Departamento de Química Analítica, Universidad de Extremadura, Av. de Elvas, s/n, 06006 Badajoz, Spain; epinilla@unex.es
2 Departamento de Física Aplicada, Universidad de Extremadura, Av. de la Universidad, s/n, 10005 Cáceres, Spain; cmiro@unex.es
3 Centro de Ciências e Tecnologias Nucleares, Estrada Nacional 10, ao km 139,7, 2695-066 Bobadela LRS, Portugal; eva.andrade@ctn.tecnico.ulisboa.pt (E.A.); mcapucho@ctn.tecnico.ulisboa.pt (M.R.); madruga@ctn.tecnico.ulisboa.pt (M.J.M.)
4 Laboratório de Proteção e Segurança Radiológica, Instituto Superior Técnico, Universidade de Lisboa, Estrada Nacional 10, ao km 139,7, 2695-066 Bobadela LRS, Portugal
* Correspondence: samfrutosp@unex.es; Tel.: +34-924-28-93-00
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Abstract: Thoron isotope has been growing in importance in the last years because, despite of its short half-life, can also contribute in the development of lung cancer. In this work, we assess the thoron massic exhalation rate and emanation factor of some building materials commonly used in the Iberian Peninsula. A continuous thoron monitor was used to measure the thoron activity growth as a function of time. To calculate the emanation factor, the activity of thoron radionuclide mother element (Th-232) by gamma spectrometry was employed. The exhalation rate values range from 0.0007 to 0.040 Bq·kg$^{-1}$·s$^{-1}$ for gypsum and Gris Quintana granite, respectively and the emanation factor range from 0.2 to 4.8% for Gran Beige and Gris Quintana granites, respectively. Ranges of results of all these parameters are in good agreement with the values reported by other authors for building materials of different countries.

Keywords: thoron; building materials; massic exhalation rate; emanation factor

1. Introduction

The radon isotopes, Rn-222 (radon) and Rn-220 (thoron) and their progenies are responsible for more than 50% of natural radiation dose and the second leading factor in the case of the development of lung cancer after smoking [1]. Both isotopes are produced from the uranium and thorium radioactive series in the earth’s crust at a comparable rate. Commonly used earth-based building materials, also contain uranium and thorium in varying amounts. The radioactive gases produced in these materials are also transported to indoor air through diffusion and convective, so building materials used for construction of houses, are considered as major sources of these gases in indoor environment. Thoron gas is part of the radioactive decay series of thorium, which is also present in building materials. The health risk from thoron (Rn-220) is usually ignored owing to its short half-life (55.6 s), but the generated thoron decay products can cause a significant dose contribution. In this work, the thoron exhalation and emanation properties were studied in building materials commonly used in the Iberian Peninsula (Portugal and Spain).
2. Materials and Methods

2.1. Materials and Sample Preparation

Samples of building materials, weighing from 1.0 to 5.0 kg, were collected from quarries and from suppliers and/or factories suppliers. The collected materials can be included in two categories: natural materials (NM) and materials incorporating residues from industries processing naturally-occurring radioactive material (PM) (Directive 2013/59/Euratom, 2014, Annex XIII) and are distributed as following:

NM: (A) Cements used in bulk amounts and superficial application (Cement of Portland Type II with fly ash of 20%. (B) Natural stones as granite and slate used both as bulk and superficial products. These samples of stones came from quarries of Extremadura region (Spain).

PM: Gypsum used in superficial application.

Building materials were crushed and dried in an oven, at 105 °C for 48 h. After that, they were grounded, sieved and the fraction less than 2 mm particle size was collected for analyses. The dried samples were put in cylindrical plastic boxes of 190 cm³ or in 1000 cm³ Marinelli beakers. In both cases, the containers were hermetically sealed for at least 28 days before proceeding to the measurement.

2.2. Determination of Radionuclide Content by Gamma Spectrometry

To obtain the Th-232 content an HPGe semiconductor detector, 45% relative efficiency at 1.33 MeV, coupled to a 4096-channel analyzer was used. The detector efficiency was calibrated using certified mixed gamma standards QCY-48, supplied by Amersham, for the energy range 60–1900 keV. Spectra were recorded during 48 h. The reliability of the spectrometer was checked by measuring the activity of an IAEA reference material, IAEA-6-SOIL. The Th-232 activity was determined by means of the γ-emissions of Ac-228 (911 keV) and Tl-208 (583.01 keV).

The activity concentration \( C_{Th} \) for Th-232 was calculated using the relation:

\[
C_{Th} = \frac{N}{t P \epsilon_f}
\]

where \( N \) is the net counts, \( t \) is the data collection time, \( P \) is the emission probability and \( \epsilon_f \) is the efficiency of the detector for the corresponding peak. The error was calculated from the combined uncertainty in the efficiency of the detector and the uncertainty in the net count rate. In this case, where several gamma-ray peaks for the calculations of the Th-232 activity are used, uncertainty in the yield is also included.

2.3. Determination of Massic Exhalation and Emanation

Exhalation is the thoron activity that diffuses per unit of time from a material into the air surrounding the material, in Bq·s⁻¹, as defined by the Netherlands Standardization Institute [2]. The exhalation can be related to the mass of the samples (massic radon exhalation, in Bq·kg⁻¹·s⁻¹). A similar method used for the Rn-222 in a previous reference was used [3]. The soil samples were put into the air-tight accumulation chamber coupled to a continuous radon/thoron monitor RTM1688-2 of SARAD (see Figure 1). The thoron emanated from the grains of the sample migrates through the pores and is finally exhaled from the surface. The equipment has an internal pump working at a flow rate of 0.30 L·min⁻¹. The thoron concentration inside the container was measured for a period of 10 days at intervals of 2 h. The detection limit is 5 Bq·m⁻³ for measurement cycles of 2 h.
The calculation of thoron exhalation was adopted from the calculation method of radon exhalation [3]. The thoron exhalation was calculated according to the following equation:

\[ E = \frac{C_{\text{thoron}} \lambda V}{M} \]  

(2)

where \( E \) is the massic thoron exhalation (Bq·kg\(^{-1}·s\(^{-1}\)), \( C_{\text{thoron}} \) (Bq·m\(^{-3}\)) is the average concentration of thoron in the container during of interval of measurement, \( \lambda \) the thoron decay constant, \( V \) (m\(^3\)) the air volume of the container and \( M \) (kg) the mass of the sample.

The emanation factor, \( \varepsilon \), representing the percentage of the produced radon from the grains that finally enter at the porous system of the sample, was calculated by the following equation [4]:

\[ \varepsilon = \frac{E}{C_{Th}} \]  

(3)

where \( C_{Th} \) is the Th-232 content (Bq·kg\(^{-1}\)).

3. Results and Discussion

The results of massic thoron exhalation, \( E \), activity concentration for Th-232, \( C_{Th} \), and thoron emanation factor, \( \varepsilon \), are summarised in Table 1.

| Category | Building Material | \( E \) (×10\(^3\) Bq·kg\(^{-1}·s\(^{-1}\)) | \( C_{Th} \) (Bq·kg\(^{-1}\)) | (%) |
|----------|------------------|---------------------------------|-----------------|-----|
| NM       | Cement           | 0.9 ± 0.4                       | 12 ± 1          | 0.6 ± 0.3 |
|          | Gran Beige granite | 1.0 ± 0.4                      | 41 ± 2          | 0.2 ± 0.1 |
|          | Gris Quintana granite | 40 ± 3                       | 67 ± 2          | 4.8 ± 0.3 |
|          | Villar del Rey slate | 5 ± 2                        | 75 ± 2          | 0.6 ± 0.1 |
| PM       | Gypsum           | 0.7 ± 0.3                       | 1.4 ± 1.4       | 4 ± 4 |

On average, the exhalation rate values range from 0.0007 to 0.040 Bq·kg\(^{-1}·s\(^{-1}\) for gypsum and Gris Quintana granite, respectively. The results show that the thoron exhalation rate is higher in granites samples and lower in gypsum samples. This can be presumably explained by the different distributions of Ra-224 mother element in the different types of samples. It should be noted how the difference among the values of exhalation rate in granites reveal their different mineralogical composition.

The highest value for activity concentration of Th-232 is shown by Villar del Rey slate (75 ± 2 Bq·kg\(^{-1}\)), being the lowest for gypsum (1.4 ± 1.4 Bq·kg\(^{-1}\)). The emanation factor range from 0.2 to 4.8% for Gran Beige and Gris Quintana granites, respectively. In the case of gypsum, owing to the low activity concentration of Th-232, the emanation data are reported with higher uncertainty. In any case the ranges of results of all these parameters are in good agreement with the values reported by other authors for building materials of different countries [5–7].
4. Conclusions

Continuous thoron monitor has been proved as a proper technique to assess some of the most important radiation properties of thoron in some building materials commonly used in the Iberian Peninsula, giving results that are in good agreement with the values reported by other works.

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Conflicts of Interest: The authors declare no conflict of interest.

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