Radiation Protection Dosimetry (2014), Vol. 160, No. 1–3, pp. 177–180
Advance Access publication 7 April 2014
doi:10.1093/rpd/ncu063

# DEVELOPMENT OF A COUPLE OF METHODS FOR MEASURING RADON EXHALATION FROM BUILDING MATERIALS COMMONLY USED IN THE IBERIAN PENINSULA

C. Miró[1], E. Andrade[2,*], M. Reis[2] and M. J. Madruga[2]
[1]Departamento de Física Aplicada, Universidad de Extremadura, Av. de la Universidad, s/n, Cáceres 10005, Spain
[2]Instituto Superior Técnico, Campus Tecnológico e Nuclear, Laboratório de Proteção e Segurança Radiológica, Estrada Nacional 10 (km 139,7), Bobadela LRS 2695-066, Portugal

*Corresponding author: eva.andrade@ctn.ist.utl.pt

**Radon is considered to be the main contributor to the worldwide population exposure to natural sources of radiation and so a lot of efforts have been made in most countries to assess indoor radon concentrations. Radon exhales from the earth's surface and is part of the radioactive decay series of uranium, which is also present in building materials. In this work, measurements of radon exhalation rates in building materials commonly used in the Iberian Peninsula have been carried out by using two different methods: active and passive techniques. In the first technique, the radon exhalation rate was measured following the radon activity growth as a function of time, by using a continuous radon monitor. The second technique is based on integrated measurements by using solid-state nuclear track detectors and a Spark Counter reading equipment. The results obtained by both measuring methods were found to be consistent.**



## INTRODUCTION

Natural radionuclides are present in the earth crust in different amounts, as well as in earth-derived building materials such as cement, bricks, sand, natural stones, etc. Building materials contribute to exposure to natural radiation by external gamma radiation from $^{40}$K, $^{226}$Ra and $^{232}$Th and by internal exposure due to the inhalation of radon and radon decay products[(1)]. Enhanced levels of those natural radionuclides in building materials may cause additional doses due to external and internal exposure[(2)]. Radon is considered to be the main contributor to the worldwide population exposure to natural sources of radiation and can cause a significant health hazard when present in high concentrations in the indoor air[(3, 4)]. Its progenies, mainly $^{214}$Po and $^{218}$Po alpha emitters, have a high damaging potential to the lung tissue and are considered to be an increasing factor to lung cancer incidence in humans. Furthermore, there is a considerable public awareness and a growing concern in relation to radon exhalation from building materials, particularly those that are used for interior decoration[(5)]. Therefore, a lot of efforts have been made in most countries to assess and control indoor radon concentrations[(6)]. To do so, it is important to evaluate the contribution of several building materials that can act as indoor radon sources in order to establish a classification of materials based on its corresponding exhalation rates or ultimately in its potential risk to radon exposure. In this work, measurements of radon exhalation rates in ornamental rocks (granites and slates) commonly used as building materials in the Iberian Peninsula dwellings and workplace have been carried out by using two different methods: active and passive techniques.

## MATERIALS AND METHODS

Two types of building materials were tested in this study. Gran Beige granites and Villar del Rey slates are extracted in the region of Extremadura (Spain), near the Portuguese border (Figure 1), and could be considered as representative of those commonly used in the Iberian Peninsula. Gran Beige granites extracted in Garrovillas de Alconétar (Cáceres) are characterised by their big beige feldspars, being widely used in historical buildings of the region as well as in other buildings in Spain and across Europe. Villar del Rey slates are characterised by the dark colour and the smooth and homogeneous surface. This material is commonly used in houses for flooring and walls cover.

In the laboratory, the samples were crashed, sieved (<2 mm), dried, homogenised and placed (a sample height of 4 cm with ∼200 g) inside 2-l (10-cm diameter per 28 cm height) cylindrical sealed containers.

By this processing, the physical characteristics influencing the radon exhalation rate substantially change (mainly due to grain sizes and humidity), meaning that the determined exhalation rate could differ from the value of the original building material. However, the main purpose of this study is to evaluate the consistency of the results obtained by using both methods.

In one of the methods (active technique), the radon exhalation rate was measured by using a continuous


Figure 1. Location of the sampling points in the Iberian Peninsula.

Figure 2. Experimental set-up for the radon exhalation measurements.

radon monitor RTM1688-2, from SARAD, in order to follow the radon activity growth as a function of time and determining the average equilibrium of the gas concentration during the exposure period (Figure 2). The radon 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 0.25 l $min^{-1}$.

The radon concentration inside the container was measured for a period of ~10 d at intervals of 2 h.

The variation of the radon concentration $C$ (Bq $m^{-3}$) inside the container can be expressed as

$$\frac{dc}{dt} = \frac{EM}{V} - \lambda C - \alpha C \qquad (1)$$

where $C$ (Bq $m^{-3}$) is the radon concentration, $E$ (Bq $kg^{-1}$ $s^{-1}$) the radon-specific exhalation rate, $M$ (kg) the mass of the sample, $V$ ($m^3$) the air volume of the container, $\lambda$ ($s^{-1}$) the radon decay constant and $\alpha$ ($s^{-1}$) the leakage rate from the container.

By solving Equation (1), the radon concentration growth as a function of time is given by

$$C(t) = \frac{EM[1 - e^{-(\lambda+\alpha)t}]}{(\lambda + \alpha)V} + C_0 e^{-(\lambda+\alpha)t} \qquad (2)$$

being $C_0$ (Bq $m^{-3}$) the radon concentration at $t = 0$.

The other method (passive technique) is based on integrated measurements by using solid-state nuclear track detectors placed on the inner upper surface of the containers. LR115 type II plastic track detectors from Kodak were used for this purpose. After 83 d of exposure, the detectors were chemically etched in an NaOH solution in a water bath at constant temperature. The resulting alpha tracks were counted using a Spark Counter model UFC-2 equipment. After obtaining the radon activity concentration, the radon-specific exhalation rate $E$ (Bq $kg^{-1}$ $h^{-1}$) can be

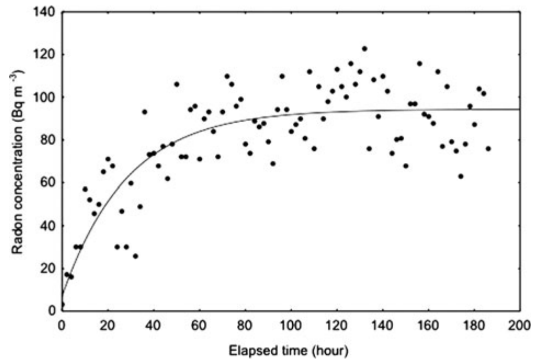


Figure 3. Growth curve of the radon concentration inside the sample container; solid line shows the fitting curve to the measured radon concentration for the Villar del Rey slates.

**Table 1. Radon-specific exhalation rate of the building materials obtained by two different methods.**

| Building material | $E$ (Bq kg$^{-1}$ h$^{-1}$) | |
|---|---|---|
| | Active technique (continuous radon monitor) | Passive technique (LR115 track detectors) |
| Gran Beige granite | 0.015 ± 0.002 | 0.011 ± 0.002 |
| Villar del Rey slate | 0.009 ± 0.003 | 0.009 ± 0.001 |

calculated through the following expression:

$$E = \frac{C_i V \lambda}{M[T + (1/\lambda)(e^{-\lambda t} - 1)]} \quad (3)$$

where $C_i$ (Bq m$^{-3}$ h$^{-1}$) is the integrated radon concentration obtained by the LR115 track detectors, $V$ (m$^3$) the volume of the container, $\lambda$ (h$^{-1}$) the radon decay constant, $T$ (h) the detector exposure time and $M$ (kg) the mass of the sample.

## RESULTS

Figure 3 shows an example of the growth curve of radon concentration due to exhalation from one of the samples under study: Villar del Rey slates.

The radon-specific exhalation rates $E$ (Bq kg$^{-1}$ h$^{-1}$) for the two types of building materials under study are presented in Table 1.

It can be observed that the granite sample (Gran Beige granite) has a higher radon-specific exhalation rate than the slate sample (Villar del Rey slate). It also can be seen that the results obtained by applying two different methods (active technique and passive technique) are consistent.

The results were compared with those measured worldwide in different types of granites and slates. The radon-specific exhalation rates for several types of granites from different origins ranges from 0.0043 to 0.238 Bq kg$^{-1}$ h$^{-1}$[7–10]. It was not possible to find in the literature the exhalation rates for slates, but the results for different kinds of ceramic tiles and ornamental rocks used for interior decoration purposes ranges from 0.001 to 0.038 Bq kg$^{-1}$ h$^{-1}$[8, 10–12].

## CONCLUSIONS

Two different methods were applied in order to determine radon exhalation rates from typical ornamental rocks (granites and slates) commonly used in the Iberian Peninsula for interior decoration purposes.

Radon-specific exhalation rates were found to be higher in granite samples, in comparison with the ones measured in slates, and are in the same range of those reported for other countries.

The results obtained through both measuring methods proved to be consistent.

Further studies are already ongoing by measuring original building materials, without previous processing, in order to determine more realistic exhalation rates.

## FUNDING

Financial support provided by Junta de Extremadura (Project PRI09A092 and FEDER-group GRU09053).

## REFERENCES


1. European Commission. *Radiological protection principles concerning the natural radioactivity of building materials.* Radiat. Prot. **112**, 16 (1999).
2. Marocchi, M., Righi, S., Bargossi, G. M. and Gasparotto, G. *Natural radionuclides content and radiological hazard of commercial ornamental stones: an integrated radiometric and mineralogical-petrographic study.* Radiat. Meas. **46**, 538–545 (2011).
3. The United Nations Scientific Committee on the Effects of Atomic Radiation. *UNSCEAR 2000 Report, Volume I: Sources.* United Nations (2000).
4. The World Health Organization. *WHO Handbook on Indoor Radon*. In: A Public Health Perspective, Zeeb, H. and Shannoun, F., Eds. (Geneva: World Health Organization) (2009). ISBN: 978-92-4-154767-3.
5. Chen, J., Rahman, N. M. and Atiya, I. A. *Radon exhalation from building materials for decorative use.* J. Environ. Radioact. **101**, 317–322 (2010).
6. Dubois, G. *An overview of radon surveys in Europe*. EUR 21892 EN, PUBSY 1429 Eur. Commiss, Directorate-General, Joint Research Centre (2005). ISBN: 92-79-01066-2.
7. Stoulos, S., Manolopoulou, M. and Papastefanou, S. *Assessment of natural radiation exposure and radon exhalation from building materials in Greece*. J. Environ. Radioact. **69**, 225–240 (2003).
8. Righi, S. and Bruzzi, L. *Natural radioactivity and radon exhalation in building materials used in Italian dwellings.* J. Environ. Radioact. **88**, 158–170 (2006).
9. Sakoda, A., Hanamoto, K., Ishimori, Y. and Nagamatsu, T. *Radioactivity and radon emanation fraction of the granites sampled at Misasa and Badgastein*. Appl. Radiat. Isot. **66**(5), 648–652 (2008).
10. Chauhan, R. P. *Radon exhalation rates from stone and soil samples of Aravali hills in India, Iran.* J. Radiat. Res. **9**(1), 57–61 (2011).
11. Gupta, M., Saini, M. and Chauhan, R. P. *Measurement of alpha radioactivity in some building construction materials.* Asian J. Chem. **21**(10), 52–55 (2009).
12. Verità, S., Righi, S., Guerra, R. and Jeyapandian, M. *Radon exhalation rates from zircon sands and ceramic tiles in Italy.* Radioprotection **44**(5), 445–451 (2009).