Radon concentration in indoor environments covered with Brazilian granites

Roberto Meigikos dos Anjos\textsuperscript{a}, Jimena Juri Ayub\textsuperscript{a,b}, Alberto Silva Cid\textsuperscript{a}, Renan Pereira Cardoso\textsuperscript{a}, Thiago Corrêa Lacerda\textsuperscript{1a}

\textsuperscript{a} LARA - Laboratório de Radioecologia, Instituto de Física, Universidade Federal Fluminense
Av. Gal Milton Tavares de Souza, s/n, Gragoatá, 24210-340 Niterói, RJ, Brazil.
E-mail: thiagol@if.uff.br

\textsuperscript{b} GEA-Instituto de Matemática Aplicada San Luis (IMASL), Universidad Nacional de San Luis, Consejo Nacional de Investigaciones Científicas y Técnicas, CCT-San Luis, Argentina.
Ej. de los Andes 950, D5700HHW San Luis, Argentina

Health hazard from radon concentrations, due to Brazilian granites used to cover the walls and floor in a typical dwelling room was assessed by indirect methods. Radon concentrations in the room were estimated by a simple mass balance equation and exhalation rates calculated from the measured values of \textsuperscript{226}Ra content in granite and the material properties. The results showed that the radon concentration in a room with adequate ventilation (0.5 h\textsuperscript{-1}) will be lower than 100 Bq m\textsuperscript{-3}, value recommended as a reference level by the World Health Organization.

XXXIV edition of the Brazilian Workshop on Nuclear Physics,
Foz de Iguacu, Parana state, Brasil
5-10 June 2011

\textsuperscript{1} Speaker
1. Introduction

Igneous rocks, such as granites, are well known to show significant quantities of radioactive isotopes in its mineral composition. Among these can be stand out $^{222}\text{Rn}$, a naturally occurring radioactive noble gas from the decay of $^{238}\text{U}$ series, with $^{226}\text{Ra}$ as its immediate parent nuclide. Radon atoms are continuously generated in the rock matrix and emanate into the air-filled pore space, from where some atoms reach the surface and escape into the atmosphere [1]. The home exposure to radon represents about half of all non medical exposure to ionizing radiation. Radon may be considered, then, a major source of radiation disease that could cause lung cancer [2].

Recently, the use of granite as building material and ornamental purposes has been popularized around the world. On the other hand, its use has caused a considerable public concern about the enhanced natural radiation exposure levels in dwellings.

In this paper indirect methods are used to predict radon concentrations in a typical dwelling room, where the walls and floor has been covered with different kinds of Brazilian commercial granites.

2. Materials and Methods

2.1 Experimental procedure

One hundred of the most popular Brazilian commercial granites were collected directly from producers States: Espírito Santo (ES), Minas Gerais (MG), Bahia (BA), Rondônia (RO), Ceará (CE), Pernambuco (PE), Goiás (GO), Rio de Janeiro (RJ), São Paulo (SP), and Paraná (PR). The obtained samples were reclassified into 71 granite kinds, in accordance to extraction site, colour and mineralogical features.

In laboratory, granite samples were ground to powder, dried, packed in cylindrical plastic containers, weighed and sealed. Activity concentrations of $^{226}\text{Ra}$ were measured by $\gamma$-spectrometry using a 55% efficiency high-purity Germanium detector (HPGe). From each granite kind, 3 to 5 samples were measured.

2.2 Theoretical approaches for estimating radon exposure

The temporal variation of $^{222}\text{Rn}$ concentration inside a room can be described by a mass balance equation. This model assumes that: (a) radon is released into the room from walls and floor; (b) radon is homogeneously mixed with the room air; (c) they not react with any substance; and (d) disappear only by ventilation and physical decay. Then, the concentration in a room can be found by [1]:

$$ \frac{\partial C_i}{\partial t} = E_s S + C_o \lambda_v - C_i (\lambda + \lambda_v) $$

(1)

where $C_i(t)$ is the radon concentration (Bq m$^{-3}$) in the room at time $t$, $E_s$ is the radon exhalation rate per unit area (Bq m$^{-2}$ h$^{-1}$), $S$ is the exhaling surface area (m$^2$), $V$ is the room volume (m$^3$), $C_o$ is the radon concentration (Bq m$^{-3}$) of the outside air, $\lambda_v$ is the air removal rate due to ventilation.
Radon concentration in indoor environments
Lacerda, T.

\( \lambda \) is the decay constant of radon \((7.54 \times 10^{-3} \text{ h}^{-1})\). The UNSCEAR \([3]\) reports \(\lambda_v\) values between 0.1 h\(^{-1}\) and 3 h\(^{-1}\) for residence. Values of \(\lambda_v < 0.1 \text{ h}^{-1}\) represent cases of extremely poor ventilation. An air exchange rate of 0.5 h\(^{-1}\) is suggested for residential mechanical ventilation systems. At the steady state, the radon concentration in the room is given by:

\[
C_{Rn} = \frac{E_x S/V + C_0 \lambda_v}{(\lambda + \lambda_v)}
\]  

(2)

From the measured values of \(^{226}\)Ra activity concentrations, \(E_x\) can be theoretically calculated as \([3]\):

\[
E_x = \frac{1}{2} A_{Ra} \lambda_{Rn} \rho \eta d
\]  

(3)

where \(A_{Ra}\) is the activity concentration of \(^{226}\)Ra \((\text{Bq kg}^{-1})\), \(\lambda_{Rn}\) is the radon decay constant \((\text{h}^{-1})\), \(\rho\) is the material density \((\text{kg m}^{-3})\), \(d\) is the wall thickness \((\text{m})\), and \(\eta\) is the emanation coefficient, i.e. the fraction of radon that reaches to the wall surface by diffusion process.

3. Data analysis and results

Figure 1 shows the frequency distribution of \(^{226}\)Ra activity concentrations from the different kinds of Brazilian commercial granites (in dry conditions). The \(^{226}\)Ra content ranged from 4.9 ± 0.6 Bq kg\(^{-1}\) to 160 ± 20 Bq kg\(^{-1}\), with geometric mean of 31.6 Bq kg\(^{-1}\) (SD Geometric = 2.3 Bq kg\(^{-1}\)). The \(^{226}\)Ra content shows therefore, variations of up to two orders of magnitude between granites of different origin. This result suggests that the indoor radon content could depend strongly on the kind of granite used for cover the room.

The indoor radon concentration was evaluated for a typical dwelling room \((5.0 \text{ m} \times 4.0 \text{ m area, 2.8 m height})\), where walls and floor are covered with Brazilian commercial granites. Taking into account that granite is commonly used in 3 cm thick tiles or slabs, the walls and floor were considered 3 cm thick granite, with a density of 2600 kg m\(^{-3}\). Al-Jarallah \([4]\) and Hassan et al. \([5]\) reported for granites, typical radon emanation coefficient \((\eta)\) in the range from < 0.025 to 0.45 Bq kg\(^{-1}\). Since the emanation rate was not experimentally measured and the reported data varied significantly, the maximum value of 0.45 was used with the aim to obtain safety assessment. The \(E_x\) values were found to be in the range from 0.65 to 21.2 Bq m\(^{-2}\) h\(^{-1}\), with geometric mean of 4.2 Bq m\(^{-2}\) h\(^{-1}\) (SD Geometric 2.4 Bq m\(^{-2}\) h\(^{-1}\)). These values are similar to reported in the literature \([4, 5]\), suggesting that this simple model can be used successfully to predict the indoor radon concentrations in dwellings, when granite rocks are used as covering material.
Radon concentration in indoor environments

Lacerda, T.

Figure 1. Frequency distribution of $^{226}\text{Ra}$ content in the analyzed granites.

In order to evaluate the radon concentration values, $C_0 = 10$ Bq m$^{-3}$ was adopted, since this value has been reported as a typical outdoor $^{222}\text{Rn}$ concentration around the world [3]. Considering that part of the room volume is occupied by furniture, we can assume that the room is a cavity with $S/V = 2.0$ m$^{-1}$. This value represents the ratio of the area covered with granite slabs, from which exhalation is taking place, to the free room volume. Two configurations of air removal rate due to ventilation were considered. For the first configuration a poor air exchange rate, with $\lambda_v = 0.1$ h$^{-1}$ was considered. For the second room configuration, a high ventilation rate was assumed; such as 0.5 h$^{-1}$, value suggested for residential mechanical ventilation systems [3].

The World Health Organization [2] has recommended that each country should set a reference level of indoor radon as low as reasonably achievable. Radon concentrations in dwellings depend on several factors such as dwelling type, design and construction, local geology, soil permeability, etc. In view of the latest scientific data on health effects of indoor radon, a reference level of 100 Bq m$^{-3}$ has been recommended by [2]. Wherever this is not possible, the chosen reference level should not exceed 300 Bq m$^{-3}$. This value represents approximately 10 mSv y$^{-1}$, according to recent calculations by the ICRP [6]. Taking into account these new findings, the ICRP [6] has therefore revised the upper value for the reference level for radon gas in dwellings from the value reported in the 2007 Recommendations of 600 Bq m$^{-3}$ to 300 Bq m$^{-3}$ [7]. Above this concentration level, an action or investigation should be conducted in order to diminish the exposure to radon.

When a poor ventilation is assumed, the steady-state indoor radon concentration ($C_{Rn}$) in our room due to exhalation from the walls and floor, are in the range from 21 to 404 Bq m$^{-3}$, with geometric mean of 91 Bq m$^{-3}$ (SD Geometric = 1.9 Bq m$^{-3}$). According to ICRP [6] and WHO [2], about 49% of Brazilian granites show radon concentrations below 100 Bq m$^{-3}$, 42% show 100 < $C_{Rn}$ < 300 Bq m$^{-3}$, and only 9% show values above 300 Bq m$^{-3}$. Thus, 91% of Brazilian granites analyzed could be used in dwellings without restrictions. Figure 2 shows $C_{Rn}$ values for each kind of Brazilian granite (black circles) when the room is poorly ventilated.
If we adopt a higher ventilation rate in our room, due to operation of mechanical ventilation systems, the $C_{Rn}$ values are in the range from 10.2 to 19.9 Bq m$^{-3}$, with geometric mean of 12.5 Bq m$^{-3}$ (SD Geometric = 1.0 Bq m$^{-3}$). In this configuration all Brazilian granites analyzed would produce radon concentrations below 100 Bq m$^{-3}$; fulfilling the WHO's recommendation [2]. Figure 2 shows, as open circles, the $C_{Rn}$ values in a room with mechanical air ventilation system.

![Figure 2](image)

**Figure 2.** Concentrations of indoor radon from a standard room (4.0 m x 5.0 m area and 2.8 m high), which walls and floor are covered Brazilian granites (3 cm thick), with poor ventilation ($\lambda_v = 0.1$ h$^{-1}$) and mechanical ventilation systems ($\lambda_v = 0.5$ h$^{-1}$).

It can be concluded, then, that under normal ventilation conditions in a room, these granites do not contribute significantly to increase the radon concentration. From this point of view, radon concentrations could be reduced in existing dwellings at moderate cost, and low concentrations can usually be ensured in new buildings at reasonable or low cost. Additionally, these results indicate that the use of granite for flooring or in countertops contributes very little to the radon concentration. Granite countertop, for example, could contribute to less than 5% of radon concentration in homes, since for this situation the ratio $S/V$ shows a very low value.

**Acknowledgements**

The authors would like to thank for the financial support to Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES), Consejo Nacional de Investigaciones Científicas y Técnicas (Conicet), Universidad Nacional de San Luis (UNSL) and Fundación Bunge y Born.
References


