Radiation Physics and Chemistry 140 (2017) 141–145

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

journal homepage: www.elsevier.com/locate/radphyschem

Radioisotopes present in building materials of workplaces

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ARTICLE INFO

Keywords:
Building materials
Radionuclides
Radon
Gamma spectroscopy
Hazard indexes

ABSTRACT

The isotope ^{222}Rn is responsible for approximately half of the effective annual dose received by the world population. The decay products of ^{222}Rn interacting with the cells of biological tissue of lungs have very high probability to induce cancer. The present survey was focused in the evaluation of activity concentration of ^{222}Rn and other radioisotopes related to the building materials at workplaces at Curitiba – Paraná State. For this purpose, the instant radon detector AlphaGUARD (Saphymo GmbH) was used to measure the average concentrations of ^{222}Rn in building materials, which were also submitted to gamma spectrometry analysis for qualitative and quantitative evaluation of the radionuclides present in samples of sand, mortar, blue crushed stone (Gneissic rock), red crushed stone (Granite), concrete and red bricks. The main radionuclides evaluated by gamma spectrometry in building material samples were ^{238}U/^{226}Ra, ^{232}Th and ^{40}K. These measurements were performed at the Laboratory of Applied Nuclear Physics of the Federal University of Technology – Paraná in collaboration with the Center of Nuclear Technology Development (CDTN – CNEN). The results of the survey present the concentration values of ^{222}Rn related to construction materials in a range from 427 ± 40.52 Bq/m³ to 2053 ± 90.06 Bq/m³. The results of gamma spectroscopy analysis show that specific activity values for the mentioned isotopes are similar to the results indicated by the literature. Nevertheless, the present survey is showing the need of further studies and indicates that building materials can contribute significantly to indoor concentration of ^{222}Rn.

1. Introduction

Radon (^{222}Rn) is a radioactive gas that can be found naturally in inhabited environments and dwellings. Its concentration and activity in air present the variation according to region and mineral composition of the soil. This radioactive gas is produced by alpha decay of radium (^{226}Ra), which belongs to the radioactive series of uranium (^{238}U).

^{222}Rn is about seven to eight times heavier than air and therefore it tends to accumulate at an altitude close to the ground level that corresponds to the air breathed by humans. The study of ^{222}Rn is important because it is a radioactive gas that emitting alpha particles (α) produce 8 radioactive successive decay products (one half undergoes alpha decay and another half is beta radioactive), which can interact with the cells of biological tissue of lungs. This radiation has very high probability to induce the lung cancer (Hopke et al., 2000).

According to the research conducted by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 2006) radon and its decay products are responsible for approximately half of the effective dose received by the population from natural radiation sources.

Typical building materials may contain significant levels of radium as well as other natural radioactive isotopes of uranium and thorium. Considering the high porosity of construction materials this may cause rather exhalation rate of ^{222}Rn and its accumulation in air of dwellings. Thus, the building materials can represent an important source of indoor radon activity (IAEA, 2003).

Materials such as cement, concrete, sand, clay bricks, granite, marble, limestone and gypsum represent a source of exposure because they contain radionuclides like potassium (^{40}K), uranium (^{238}U), thorium (^{232}Th) and its decay products (Fathivand et al., 2006; Turhan et al., 2008; Mehdizadeh et al., 2011).

According to the European Commission (1999), when an individual inhabits an environment built from concrete blocks that contain an average concentrations of radium activity, thorium and potassium

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http://dx.doi.org/10.1016/j.radphyschem.2017.02.037
Received 3 October 2016; Received in revised form 16 February 2017; Accepted 17 February 2017
Available online 20 February 2017
0969-806X/

equal 40 Bq/kg, 30 Bq/kg and 400 Bq/kg, respectively, this will cause an increase of his effective annual dose of about 0.25 mSv/year.

International Agencies such as the Environmental Protection Agency of the United States (USEPA, 2016) and the International Commission on Radiological Protection (ICRP, 1993, 2010, 2014) have elaborated guides of protection measures and radon prevention as well as recommendations for reduction of radon levels in air of residential areas and workplaces.

The ICRP states that the value of 300 Bq/m^3 meets the basic standards of safety standards recommended by agencies such as the International Atomic Energy Agency (IAEA), United Nations Environment Programme (UNEP), World Health Organization (WHO), The European Atomic Agency Community (EURATOM), among others.

The Commission also states that the radon reference limits should be applied in mixed-use buildings that are used both by individuals and by the public workers (ICRP, 2014).

Radon surveys concluded in different regions of Brazil present results that confirm rather high concentrations of ^{222}Rn in air of dwellings (Corrêa, 2006; Veiga et al., 2003; Geraldo et al., 2005; Neman, 2000; Santos, 2010), which in some cases reaches the values from 200 to 600 Bq/m^3. During the last years, an increase of experimental studies concerning ^{222}Rn concentration in air of Brazilian homes and dwellings was observed. Unfortunately, it is not sufficient yet for the evaluation of the overall population exposure to this sort of radiation.

The aim of the present research was to study the correlation between the radon activity in air of workplaces and concentration of other radionuclides present in collected samples of building materials from those locations.

2. Materials and methods

The present survey was focused in the evaluation of activities concentration of ^{222}Rn and other radioisotopes related to the building materials at workplaces at Curitiba – Paraná State (Brazil), which is a big industrial city located in Southern Brazil. The urban area of the city is situated at the First Paraná Plateau located between the Serra do Mar Mountain and Devonian Cliff which makes part of the hydrographic basin of Alto Iguaçu River. The substrate soil of Paraná, contains rocks, which are mostly classified as metamorphic. At this region a great variety of igneous intrusive rocks could be found, including granites and granitoids (MINEROPAR, 2001; Pereira et al., 2013; Salamuni et al., 2003). This fact leads to the conclusion that building materials from this region could contribute considerably to the concentration of radon in air of dwellings and workplaces of this region since granites usually contain significant amount of the precursor (parent) radioisotopes as uranium and thorium.

The samples of sand, mortar, blue crushed stone (Gneissic rock, mesh size of approximately 30 mm), red crushed stone (Granite, mesh size of approximately 38 mm), crushed concrete and red bricks (characteristic size of 80 mm) were randomly chosen and separated from different construction places of Curitiba metropolitan area, mainly from recently build new academic sciences, research and laboratory buildings of the Federal University of Technology – Paraná (UTFPR). All samples were submitted to analysis concerning the exhalation of ^{222}Rn as well as the evaluation of radionuclides such as ^{226}Ra, ^{232}Th and ^{40}K, which are the main elements associated with human exposure to radiation from building materials.

From collected building materials samples of 1 kg were separated and stored in sealed in glass vessels (Fig. 1) for 40 days while ^{222}Rn reaches the secular equilibrium. Accumulated radon activity in air of sealed bottles was measured using the instant radon detector AlphaGUARD (Saphymo GmbH).

For the purpose of background evaluation, an empty glass vessel

Fig. 1. Brick sample stored in a glass vessel to measure the ^{222}Rn concentration.

was separated and used as reference in each measurement being stored at the same conditions as the glass vessels with material samples. Filters, hose and connectors were added in the air circuit measurement as shown in Fig. 2. All components and connections were tested for possible points of entry of external air as well as leakage of accumulated gas that could alternate the results obtained with the AlphaGUARD.

Every measurement consists of three major steps: (1) AlphaGUARD ventilation using the open circuit, (2) background measurements, and (3) measurements of ^{222}Rn liberated by building material sample and accumulated within the internal volume of the sealed bottles. All steps were performed using the air pump (AlphaPUMP), supplied together with the AlphaGUARD, which operated with air flow rate of 0.5 L/min. The AlphaGUARD was set to operate in mode of "1 min/flow" suggested by the User Manual.

The ventilation of the AlphaGUARD was done using open-air circuit when the air pump remained turned on for 40 min approximately, so that the detected alpha activity reaches the lowest level before the background evaluation step (Fig. 3).

Background measurements were performed during 1 h using a closed air circuit that prevents the entry of external air into measurement system. In this second step the air circulation circuit contains the precision rotameter, two dust filters and safety glass vessel that is included in Aquakit of AlphaGUARD system. All those elements were used in the measurements in order to ensure that possible dust particles from the samples will not penetrate inside the ionization chamber of AlphaGUARD.

At the third step, the glass vessels with building materials samples were connected to the AlphaGUARD and submitted to evaluation during two hours. This step was executed using the air pump operated with airflow rate of 0.5 L/min. The AlphaGUARD detector was set to operate in mode of "1 min/flow" following the recommendations of the User Manual. Every step of materials evaluation was concluded by background control measurement to ensure that there was no contamination of the AlphaGUARD by any particles or dust from the samples.

The volume of each sample of building materials, because of its irregular form, was found using the Archimedes' principle by measuring their weight with a precision-scale in air and when they were immersed in water.

Other samples of the same building materials were sent to the Laboratory of Natural Radioactivity of the Center of Nuclear Technology Development (CDTN) for qualitative and quantitative assessment of natural radionuclides using gamma spectrometry. This analysis was performed using a gamma spectroscopy system (CANBERRA) with hyper pure germanium semiconductor detector (HPGe) solid state with high efficiency (15%) and resolution, located

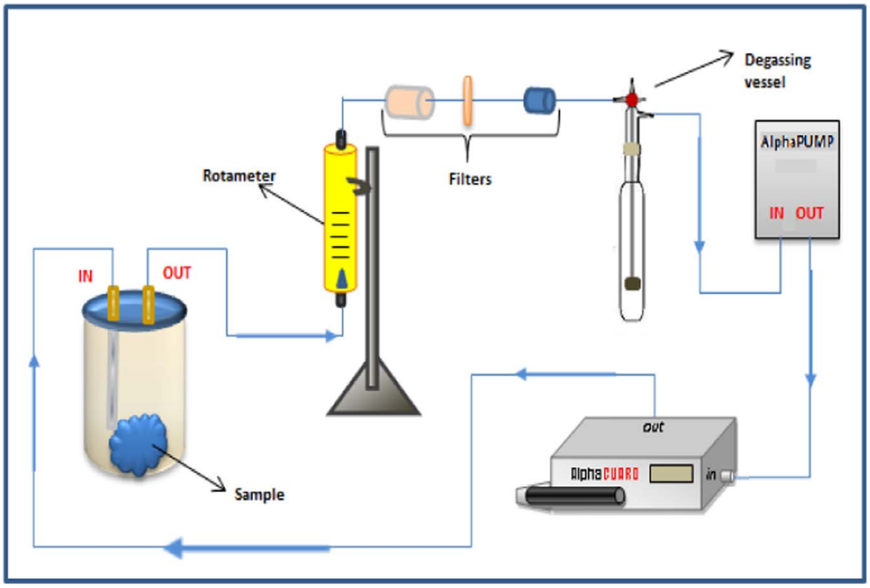

Fig. 2. Schematic diagram of the closed circuit for the measurement of background samples and building materials.

inside the Lead Shield Chamber installed and operated inside a shielded room designed for tritium analysis and measurements. Thus, the external radiation does not interfere with the equipment detection limits.

For this analysis the samples of each studied building materials were ground in a ball mill device (Los Angeles Abrasion Test, mesh size of 0.074 mm), homogenized and stored in Marinelli Beakers and remained at rest for approximately 40 days to achieve the secular equilibrium between ^{226}Ra and its decay products.

The use of Marinelli beaker is essential because they ensure that the sample remains as close as possible to the HPGe detector thereby improving the detection efficiency.

Table 1 shows the gamma transitions of ^{214}Pb, ^{214}Bi, ^{212}Pb, ^{212}Bi, ^{228}Ac, and ^{40}K, which were used in the gamma spectrometry analysis for determining the activity of ^{226}Ra, ^{232}Th, and ^{40}K in the studied building material samples.

The method of gamma spectrometry allowed the identification of radionuclides originated from the decay series of ^{238}U and ^{232}Th. Among the main natural radionuclides found in the building materials samples, there were considered the ^{226}Ra, ^{232}Th and ^{40}K isotopes, which had their higher activity (Bq/kg) (Table 2).

Table 1
Gamma transitions used in gamma spectrometry analysis.

Isotope	Gamma Transition
^{214}Pb	295.21 keV and 351.92 keV
^{214}Bi	609.3 keV
^{212}Pb	238.6 keV and 300.1 keV
^{212}Bi	727.3 keV
^{228}Ac	911.1 keV and 968.9 keV
^{40}K	1460.7 keV

Those data were used for the calculation of the concentration index of activity (I), radium equivalent (Ra_{eq}) and emanation coefficient (f) given by Eqs. (1)–(3) that are shown below:

$$I = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000}; \quad (1)$$

$$Ra_{eq} = C_{Ra} + C_{Th}\bullet 1.43 + C_K\bullet 0.077; \quad (2)$$

$$f = \frac{C_{Rn}\bullet V}{C_{Ra}\bullet M}; \quad (3)$$

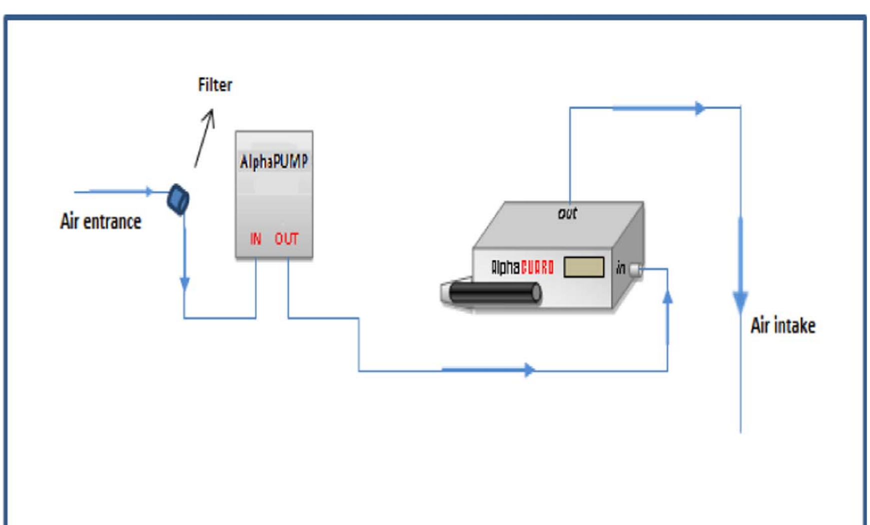

Fig. 3. Schematic drawing of AlphaGUARD ventilation in open circuit.

Table 2
Activity in [Bq/kg] of radionuclides evaluated by the technique of gamma spectrometry.

Sample	^{238}U Series	^{232}Th Series	^{40}K
	^{214}Bi (^{226}Ra)	^{212}Pb (^{224}Ra) and ^{228}Ac	
Sand	11.4 ± 0.2	12.2 ± 0.1	620.2 ± 4.5
Blue crushed stone	22.0 ± 0.3	48.8 ± 0.3	1091.6 ± 5.6
Red crushed stone	36.4 ± 0.4	66.4 ± 0.3	1058.3 ± 5.6
Mortar	10.8 ± 0.2	15.0 ± 0.1	79.1 ± 1.6
Concrete Blocks	20.4 ± 0.3	34.5 ± 0.2	849.5 ± 5.2
Red Bricks	29.9 ± 0.4	36.5 ± 0.3	178.6 ± 3.0

Table 3
Results of ^{222}Rn measurements for six studied building materials.

Material Sample	^{222}Rn Activity Concentration, [Bq/m^3]
Sand	427.0 ± 40.52
Blue Crushed Stone	500.0 ± 44.9
Red Crushed Stone	2053.0 ± 90.06
Mortar	510.0 ± 41.94
Concrete Blocks	795.0 ± 53.16
Red Bricks	1270.0 ± 66.97

In the equations above, the activity concentration index (I) was calculated following the recommendations of the European Commission (1999), where the terms C_{Ra}, C_{Th} and C_K represent the concentrations of activity of ^{226}Ra, ^{232}Th and ^{40}K in Bq/kg, respectively. The maximum value of Ra_{eq} was considered equal to 370 Bq/kg (indoor reference limit) established by international norms and the recommendations of other publications (OECD, 1979; Beretka and Mathew, 1985; Malanca et al., 1993; Venturini and Nisti, 1997; Al-Sulaiti et al., 2008; Trevisi et al., 2012)

The radon emanation coefficient (f), showed in Eq. (3), was used to evaluate the fraction of radon gas produced by radium decay in each sample of the studied building materials and liberated into the air of external environment. This coefficient was calculated using the results of measured radon equilibrium activity (C_{Rn}), the free volume of used glass vessels (V), radium activity (C_{Ra}) in studied building materials samples (Bq/kg) provided by the analysis of gamma spectroscopy, the sample mass (M) and following the approach described in (Morawska, 1989; Kovler et al., 2005; Corrêa, 2006; Bikit et al., 2011; Hassan et al., 2011; Hassan, 2014).

3. Results and discussion

The activity concentrations of ^{222}Rn exhale from building materials was obtained by subtracting the background values measured by the AlphaGUARD. The errors associated were obtained by calculating the quadratic sum of the errors associated with the background measurements and experimental ^{222}Rn activity detected by AlphaGUARD.

Table 3 presents the obtained results for radon concentration activities in the air for the studied samples of sand, mortar, blue crushed stone, red crushed stone, crushed concrete and red bricks.

Thus, the concentration levels of ^{222}Rn associated to evaluated building materials were obtained in a range from 427 ± 40.52 Bq/m³ to 2053 ± 90.06 Bq/m³, which are well above the range of activity of 10 Bq/m^3 to 20 Bq/m^3, which is established by the European Commission (1999). Such elevated levels of radon exhalation by the studied building materials showed the possibility that they can contribute potentially to the elevated indoor ^{222}Rn activity above the limit of 200 Bq/m^3 established by UNSCEAR United Nations (2000) and 300 Bq/m^3 established by the International Commission on Radiological Protection (ICRP, 2014).

It's possible to see a rather strong correlation between radon exhalation and radium content in the studied samples of building materials used in construction of workplaces at the Curitiba metropolitan area. The calculated Pearson's correlation coefficient for this pair of isotopes is 0.91.

The obtained values of the radon emanation coefficient (f) ranged from 0.9% for blue crushed stone (Gneissic rock) to 2.2% for red crushed stone (Granite). These data may be considered within the normal range according the literature (Lee et al., 2001; Al-Jarallah et al., 2005; Bikit et al., 2011), since the values of *f* normally tend to vary from 1% to 30%, depending on the building material.

Calculated values of Ra_{eq} for all samples of building materials were found below the maximum permissible value of 370 Bq/kg suggested in the literature (Al-Sulaiti et al., 2008; Beretka and Mathew, 1985; Mehdizadeh et al., 2011).

Whereas the values of activity concentration index (I) calculated based on experimental results of gamma spectrometry for the samples of sand, red crushed stone and red clay bricks were found below of 0.5, it is possible to conclude that these materials may contribute to the annual effective dose of about 0.3 mSv following the documents of the European Commission (1999). In the case of other evaluated samples of mortar, blue crushed stone and concrete clocks, the activity concentration indexes presented the values close to 1.0 that may be associated with the effective annual dose contribution of less than 1 mSv/year.

4. Conclusions

The present research presents the results of survey of radioactivity of building materials common in the Paraná State of Brazil. The obtained values of Hazard Indexes show that the activity concentrations were within the normal limits, since no value exceeded the activity limits recommended by the European Commission, ICRP, USEPA and UNSCEAR. Nevertheless, observing the rather small quantity of similar studies of Brazilian building materials and the present grow of building materials industry, and considering the absence of national norms and regulation, the importance of further studies of radioactive safety of building materials to ensure the reduction of human exposure can be highlighted.

Acknowledgments

The authors are very thankful to CNEN, CAPES, CNPq and Fundação Araucaria of Paraná State for financial support of current research work as well as to colleagues from the Center of Nuclear Technology Development (CDTN/CNEN) for the positive discussions and assistance in the measurements.

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