EVALUATION THE TECHNOLOGICAL CHARACTERISTICS OF ORNAMENTAL ROCKS IN THE EMANATION OF RADIOACTIVE GASES

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Rio de Janeiro
Julho/2012

CAC-0007-00-12 – Contribuição técnica realizada para o Global Stone Congress, realizado em Borba – Portugal, de 16 a 20 de julho de 2012.
Abstract. Due to international concern about the radioactivity in ornamental rocks, was ordered to the assistance of the Centre for Mineral Technology (CETEM) and the Institute of Radiation Protection and Dosimetry (IRD) to carry out the assessment of levels Brazilian ornamental stones radiation used as coating material. This desire is due to the alleged undue exposure to radioactive gases, caused by the exhalation of radon (\(^{222}\)Rn) granite which increases the risk to the health of living beings. On this basis, the aim of this study is to evaluate the rate of exhalation of radioactive gases in the floor boards, using nuclear techniques and the use of model calculations respected internationally accepted. Therefore, we performed radiometric and mineralogical characterization of the different types of silicate rocks used for the coating, the determination of tariffs and the exhalation of radon activity concentration and potassium in the materials chosen and these are correlated with rates the physics literature.

Keywords: ornamental stone, releases radioactive radon.

1. INTRODUCTION

The industrial sector of dimension stone of Brazil produces a large variety of granites, marbles, etc., reaching for about 500 different types of rocks. The dimension stone are usually classified in the following way: trade names, producing state, origins geological and mineralogical composition; however, are not classified as the concentration of radioactive substances. The study of the concentrations of radionuclide isolation in granitic rocks and important due to two reasons: because it is an important tool technique, given that these concentrations can be used for the petrographic classification of granite, and due to problems radiological, since the presence of the radioactive elements provides an increase in the levels of radiation present in the civil construction [1].

1.1. Radioactivity in construction materials

It is safe to say that the field of gamma radiation inside a residence, coming building materials, it is originated, mainly, by decay of radionuclides from the chains of \(^{238}\)U and \(^{232}\)Th, in addition to the \(^{40}\)K [2]. The gamma radiation, which results from the decay of natural radionuclides that are present on the materials used in building, it is generated of form isotropic and, consequently, with this, only
a fraction of the emissions originals should probably reach the exposed individual inside the building [3].

1.2. Radon
Radon (Rn) is a colorless gas, odourless and tasteless, in addition to 7.58 times heavier than air and more than a hundred times heavier than hydrogen natural, originates from the radio, an element of uranium and thorium decay series. It is present in virtually all the places of the earth's crust, and for being a gas, has the property of accumulating in closed environments such as residences, buildings, caves, mines and tunnels.

1.3. Radon - 222
The isotope $^{222}\text{Rn}$ is an alpha emitter ($T_{1/2} = 3.82$ days) and, together with his sons not gaseous $^{218}\text{Po}$ and $^{214}\text{Po}$, are responsible for approximately 50% of the effective dose equivalent produced by natural ionizing radiation. The gas radon-222 is completely natural and if form itself during the decay of uranium-238 [4].

1.4. Radon – 219 and Radon - 220
There is also the $^{219}\text{Rn}$ and $^{220}\text{Rn}$, which are products of series of decay of $^{235}\text{U}$ and $^{232}\text{Th}$, respectively. They have a long half-life very small when compared to $^{222}\text{Rn}$, being the of $^{219}\text{Rn}$ in 3.96 seconds and the $^{220}\text{Rn}$ of 55.6 seconds, thus, the radon-219 and radon-220 are not able to migrate inside the houses and generate preoccupation in the area of health, unlike the radon-222 [5]. In this work, there is no assessment of the rate of emission of radon-219, by difficult to obtain accurate data since its a half-life time is very small, compared with the other isotopes of radon. And then, it was evaluated only the radon-222, the more harmful, and the radon-220, that even having a half-life short can still be considered dangerous.

1.5. Potassium
It is important to emphasize that the natural potassium is a mixture of three isotopes, $^{39}\text{K}$, $^{40}\text{K}$ and $^{41}\text{K}$. Of the three natural isotopes, only potassium 40 (40K) is radioactive, that is an unstable nucleus, and occurs in a proportion of 0.0118 % (in grams) in relation to the total mass of potassium. The physical-chemical characteristics and properties of emission of radioactive $^{40}\text{K}$ allow its determination through techniques of direct measurements in units called counters of the whole body, these units equipped with instrumentation capable of detecting the presence of radioactive elements in living beings [6].
1.6. Activity of Sample

The unstable nuclei of the same species (even chemical element) and of different mass, called radioisotopes are not all the changes at the same time. The emissions of radiation are made so unexpected and not if you can guess the moment in which a given nucleus will emit radiation. However, for the large quantity of atoms in a sample is reasonable to expect a certain number of emissions or transformations in each second. This "rate" of transformations is called activity of the sample.

The activity of a sample with radioactive atoms (or radioactive source) is measured in Bq - Becquerel (one disintegration per second) or Ci (Curie), which is equivalent to $3.7 \times 10^{10}$ Bq.

The indices of activities serve to countersign the materials of building in terms of safety in relation to the activity, for use in building, highways and bridges. These indices are calculated on basis of the concentrations of elements $^{226}$Ra, $^{232}$Th and $^{40}$K, according to the Eq. 1 [7]:

$$I = \frac{C_{Ra}}{200} + \frac{C_{Th}}{200} + \frac{C_{K}}{8000}$$  \hspace{1cm} (1)

with the final product expressed in Bq.kg⁻¹.

Where the values of the activity indice given by equation 5 is $I = 2$ (corresponding to an annual dose of 0.3 mSv), while $I = 6$ (corresponding to a dose of up to 1 mSv/year). This way, this activity indice can be used as a tool for identifying materials that can produce changes in the level of background radiation. According to this criterion of dose, with materials $I > 6$ should be avoided, since these values will cause an increase in the rate of annual dose, above the level of background radiation, more than 1 mSv. This is the limit value, internationally recommended for the general population [8].

1.7. Physical indices

Physical indices are important tools that provide a notion of micro irregularities present in the rock, allowing indirectly evaluate the condition of change and cohesion of the rocks, and can be interpreted in accordance with the density (high-density, high porosity, high absorption and increase the saturation).

Porosity is the percentage of spaces between the clasts or microfractures in a rock or sediment. And is given by the ratio between pore volume (or empty volume) by the total volume of rock. The porosity can be intergranular primary and secondary fracture in grain and fracture in the rock. The
porosity is directly linked to the emission of radioactive gases, and the greater the porosity, the higher the rate of emission of the gas in the rocks.

2. Objective

This way, the study aims evaluation the rate of emission of radioactive gases: radon and potassium, in dimension stone brazilian. Also, we were calculated the rate of radioactive activity in closed environments, which enabled us to identify the real threat of samples with a view, the great importance of these results for the health of living beings.

3. Materials and Methods

In this work, we were analyzed three types of samples granites, known commercially as: Crema Bordeaux, Mombasa and Golden. These were selected due to the high rate of export for the use of these granites in civil construction international. The samples were found to be in the form of plates (15 cm x 30 cm x 02 cm) polished and without resin. All the samples came from region of Cachoeiro de Itapemirim, Espirito Santo state, Brazil. Two types of studies were carry out to evaluation the rate of emission of radioactive gases in these samples, being a necessary preparation of samples and another using the rocks in their natural state.

3.1. Rocks in natural state

In this study, in that, was not necessary any preparation of the sample, it is consisted of a chamber of stainless steel with 20L and removable covers sealed for retention of gas. The caps were previously tested for which there is no leakage. In this work, were put together, two plates of dimension stone, with the measures already mentioned earlier, that were left in this system for seven to eight days. The gas formed was injected into an analyzer of radon gas called Alpha Guard 2000 PRQ (Genitron Instruments) in cycles of one to two hours. During the evaluation period, set itself a microfan, which it was inside the chamber, for which there was the circulation of air inside and thus a better observation in measure of the gas by the tool used. The growth of the activity of radon in the chamber allowed estimating the value of final activity for a period corresponding to the seven half-lives of radon.

3.2. Rocks powdered

For the study in which it was necessary the preparation of the samples we used a spectrum analysis-range. For that, it was made crushing, grinding and sieving cleaning. In this work, the plates of the samples were crushed and taken to the grinding, having the conversion the dust or
spray with the aid of a sprayer (Fritsch). The sprayer was used with caution, because the samples should, in its majority, it is between 0.177 mm to 0.149 mm after the screening to obtain a better efficiency in the analysis spectrum range. Soon, it was put small quantities of the sample in the container the sprayer (made of titanium to avoid any type of contamination of samples), in 400 rpm for 2 minutes. Once removed, the ground samples were placed in a stack of sieves of 2.360 mm (to remove the balls of titanium sparge), 0.177 mm and 0.149 mm, respectively, and taken to the Rotap, appliance of agitation, where they remained for about 10 minutes, for which there was total separation of the sample in the sieves.

Despite the preference of samples between 0.177 mm to 0.149 mm, were also used other samples that were below of 0.149 mm. Before mixing and mix these, was made sieve analysis, which calculated the percentage of how much it was above or below 0.149 mm. After sieving, the samples were ready for the analysis spectrum-range. These were packed in containers, where they remained at rest for 30 days before being measured in detector (necessary to achieve the balance secular). Employees were detectors of gamma spectrometry of high purity (Germanium HPGe Detector), which allowed the identification of the emission range of $^{40}\text{K}$ and of the sons of series of U and Th. The counting time for determining the concentration of activity of these nuclides in Bq/g, varies from eight to 16 hours, according to the activity of the material. Thus, it was possible to compare the results obtained with the exclusion limit recommended by the International Atomic Energy Agency (IAEA).

4. Results and Discussions

4.1. Sieve analysis

Table 1 shows the result of sieve analysis made in the samples of Crema Bordeaux, Mombasa and Golden, respectively. Due to the difficulty of controlling the degree of spraying of the rocks it was observed that the greater part of sieve fraction was lower than 0.149 mm in all samples.

<table>
<thead>
<tr>
<th>Sieve Fraction (mm)</th>
<th>Crema Bordeaux Mass (%)</th>
<th>Mombassa Mass (%)</th>
<th>Golden Mass (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>+ 0,177</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>- 0,177 + 0,149</td>
<td>10,29</td>
<td>16,52</td>
<td>10,59</td>
</tr>
<tr>
<td>-0,149</td>
<td>89,71</td>
<td>83,48</td>
<td>89,41</td>
</tr>
<tr>
<td>Total</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
</tbody>
</table>
4.2. Emission rate of $^{222}\text{Rn}$ Gas

The Fig. 1 represents the results of the evaluating test of the emission rate of radon gas in the samples in its natural state, and may be observed that the rates for samples Crema Bordeaux, Mombasa and Golden were very low, $3.10 \times 10^{-3}$ (Bq.m$^{-2}$), $1.60 \times 10^{-3}$ (Bq.m$^{-2}$), $6.84 \times 10^{-4}$ (Bq.m$^{-2}$), respectively.

![Figure 1: Rate of emission of radon in each sample in its natural state](image)

4.3. Radiometric Analysis / radiochemical

The determinations of $^{226}\text{Ra}$, $^{40}\text{K}$, $^{232}\text{Th}$ and Range were conducted after the balance of radionuclides (30 days) and with a detector of germanium, as stated before in the methodology. The measurements were made by counting alpha and beta, in a proportional detector low background, after chemical separations and the measures of thorium-232 by spectrometry with arsenazo III. All results are in the unit Bq.kg$^{-1}$.

Exposure to radiation, mainly due to the following radionuclides determined in Fig. 2. Where, of the elements of $^{232}\text{Th}$ and $^{226}\text{Ra}$ are formed by their decay series, the gases radioactive $^{220}\text{Rn}$, $^{222}\text{Rn}$, respectively. It is important to remember that the $^{40}\text{K}$, higher rate of emission of gamma rays in all samples analyzed, does not belong in the decay series of any kind of radon.

![Figure 2. Main Radionuclide](image)
4.4. Activity indices
It was possible to calculate the rate of activity of the samples with the aid of equation 3, described in the introduction.

- For the sample of Crema Bordeaux:

It is known that the concentrations of $^{232}$Th (Cth), $^{226}$Ra (CRA), $^{40}$K (CK) are, respectively, equal to 6.38 E1, 4.48 E2 and 1.14 E3. Then, can the activity indices (I) shall be equal to:

\[ I = \frac{6.38 \times 10^1}{200} + \frac{4.48 \times 10^2}{300} + \frac{1.14 \times 10^3}{3000} \]
\[ I = 3.19 \times 10^{-1} + 1.493 \times 10^0 + 3.80 \times 10^{-1} \]
\[ I = 2.189 \times 10^0 \]

- For the sample of Mombassa

To the concentrations of $^{232}$Th (Cth), $^{226}$Ra (CRA), $^{40}$K (CK) are, respectively, equal to 3.44 E1, 1.93E2 and 1.181 E3. Then, can the activity indices (I) shall be equal to:

\[ I = \frac{3.44 \times 10^1}{200} + \frac{1.93 \times 10^2}{300} + \frac{1.181 \times 10^3}{3000} \]
\[ I = 1.72 \times 10^{-1} + 6.43 \times 10^{-1} + 3.936 \times 10^{-1} \]
\[ I = 1.209 \times 10^0 \]

- For the sample of Goklen:

It is known that the concentrations of $^{232}$Th (Cth), $^{226}$Ra (CRA), $^{40}$K (CK) are, respectively, equal to 2.77 E1, 9.96 E1 and 1.19 E3. Then, can the activity indices (I) shall be equal to:

\[ I = \frac{2.77 \times 10^1}{200} + \frac{9.96 \times 10^1}{300} + \frac{1.19 \times 10^3}{3000} \]
\[ I = 1.385 \times 10^{-1} + 3.32 \times 10^{-1} + 3.967 \times 10^{-1} \]
\[ I = 8.672 \times 10^{-1} \]

5. Conclusion

According to the results of this work, it was observed that the rate of exhalation of radon gas in the samples analyzed was very small. Having the $^{222}$Rn, radionuclides most dangerous to human health, presented lower rates of exhalation of all types of radon. Already the $^{40}$K, had the highest rate of emission of gamma rays in all samples analyzed, i.e. he is the element which has the greatest energy peak. Thus, the most dangerous to human health, as it is this energy that can cause damage and/or mutations in cells of the interior of the human body. You can also observe that the physical parameter such as porosity, not significantly interferes with the rate of exhalation among the granite, which is very low for this type of rocks. Apparently, the granite Mombasa was the
discrepancy in the parameter of the correlations when compared with the other studied. Thus, we can conclude that the granites analyzed in this study were not alarming levels in terms of the exhalation of radon gas and potash.

6. References


