EVALUATION OF RADON´S EMISSION IN ORNAMENTAL ROCKS

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Abstract

The representative organizations of the sector of ornamental rocks had looked the Commission of Nuclear Energy by means of the Center of Mineral Technology - Espirito Santo (CETEM-ES) requesting aid how much the evaluation of levels of radiation in Brazilian ornamental rocks, exported to U.S.A. for use as material coverings in residences. Exhalation of radon (Rn) from these exotic granites may expose the occupants to an increased risk of contracting lung cancer. The objective of this work was to evaluate the risk associated with the exhalation of radon of indoor covering plates in environments, by means of nuclear techniques and to use models of calculation of dose endorsed internationally. For in such a way, the characterizations radiometric and mineralogical of three types of silicatic rocks used for covering have been carried through, determining the rates of exhalation and concentration of radon´s activity in the chosen materials.

Introduction

The Brazilian industrial sector produces a great variety of granites, marbles, quartzite etc., reaching to all about 500 different types of rocks. Normally, these ornamental rocks are classified by corporate names, the producing state, the geologic origins and mineralogical composition. However, they are not classified by radiological activity. The study of radioelement´s concentrations in granites rocks is important for the classification granite petrography for application in civil construction. Granites are an example of rocks that contain a natural enrichment of uranium - 238 (^{238}U); thorium - 232 (^{232}Th) and potassium - 40 (^{40}K). The concentrations of these elements in the rock vary significantly from sample to sample. However studies have shown that the general petrography characteristic is closely correlated to the concentration of these radioactive elements (Whitfiel et al. 1959). In terms of surface mobility, uranium and potassium can easily be oxidized into water soluble compounds that can be leached from the surface matrix and deposited in sediments far from the origin of the rock. On the other hand, Thorium, which is less water soluble, is less susceptible to leaching and tends to stay within the rock matrix. Thus, over time this difference in solubility results

in the observed petrography characteristics of granite rocks and produce specific alterations in the ratios of natural radionuclides concentration: Th, U, K, Th/U and Th/K (Anjos *et al,* 2004).

1.1. Radioactivity in construction materials

It is generally accepted that indoor residential gamma radiation exposure from construction materials comes primarily from the decay of ^{238}U and ^{232}Th, beyond ^{40}K and their subsequent daughters.. For example in the ^{238}U and ^{232}Th decay´s series there are 16 daughters that release gamma radiation with emission greater that 10^{-3} fotons (Pink, 1997). However, only a small fraction of this gamma radiation actually results in dose to the occupant (Fernandes *et al*, 2004).

1.2. Radon in residences

The primary means of radon entrance into residences is by means of cracks around foundations and drains, diffusion through floor coverings and other openings in the floor. Consequently, the Office of the General Surgeon and the Environmental Protection Agency (USEPA) recommend that radon tests be performed in all houses. In addition, radon exposure from water ingestion, in combination with radon inhalation, can also represent a significant risk to the population. This exposure combination exposes sensible cells in the respiratory and gastrointestinal tract results in increased risk of different types of cancers. However, the greater risk is to the lungs resulting in about 10,000 American deaths per year (HyperPhysics, 2009).

Because granites are commonly used throughout the world in homes as ornamentation accents, counter tops and other interior coverings, radiation exposure concerns raised by the media gave rise to many baseless rumors about the overall safety of Brazilian granites. Consequently, Brazilian granite commercialization was impacted in some countries in Europe and Asia. Therefore, the relevance of this scientific study to address these misconceptions is both timely and important to Brazil.

1.3. Radon

Radon is a colorless, odorless. tasteless gas, that is 7.58 times heavier than air and more than one hundred times heavier than hydrogen. It originates from either radium or thorium and is a member series of uranium decay series. These precursors of radon are found throughout the terrestrial crust, therefore all homes worldwide are potentially at risk. By virtue of being the only gas in the uranium decay series, radon has the potential to migrate from the soil or rock matrix and accumulating in residences, caves, mines and tunnels. The isotope ^{222}Rn is an alpha emitter ($T_{1/2}$ = 3,82 days), together with its not gaseous daughters ^{218}Po and ^{214}Po, are responsible for approximately 50% of the dose from natural ionizing radiation. Other isotopes of radon include ^{219}Rn and ^{220}Rn, which are products of ^{235}U and ^{232}Th, respectively. However, they have the very short half-life when compared with ^{222}Rn, being of ^{219}Rn of 3.96 seconds and of ^{220}Rn of 55.6 seconds. Consequently Radon-222 is the only isotope capable of migration from the

soil and rock matrixes into homes (Chyi, 2008). Table 1.1, shows the Uranium-238 decay series (Craig, 2008).

Table 1.1 Decay of uranium-238

ORIGINAL ELEMENT	TRANSFORMED ELEMENT	TIME OF HALF-LIFE	EMISSION
Uranium - 238	ThoriumThorium - 234	4,5 trillions of years	One alpha particle
ThoriumThorium - 234	Protactinium – 234	24,5 days	One beta particle and one gamma ray
Protactinium-234	Thorium – 230	269 thousandth years	One beta particle and one gamma ray
Thorium - 230	RadiumRadium – 226	83 thousandth years	One beta particle and one gamma ray
Radium - 226	Radon - 222	1590 years	One beta particle and one gamma ray
Radon - 222 *	Polonium– 218	3,925 days	One alpha particle
Polonium - 218	Lead – 214	3,05 minutes	One alpha particle
Lead - 214	Bismuth – 214	26,8 minutes	One beta particle and one gamma ray
Bismuth - 214	Lead-210 to Polonium-214	19,7 minutes	One alpha particle or one beta particle and one gamma ray
Polonium - 214	Lead – 210	150 micro second	One alpha particle
Lead - 210	Lead – 210	1,32 minutes	One beta particle
Lead - 210	Bismuth – 210	22 years	One beta particle and one gamma ray
Bismuth - 210	Polonium - 210	5 days	One alpha particle and one gamma ray
Polonium - 210	Lead - 206 **	138 days	One alpha particle and one gamma ray

** Is a steady isotope of lead.

1.4. Acceptable Limits of Gas Radon

The primary dose from Radon and its daughters occurs from inhalation, which deposits its energy in the lungs. However, in accordance with the effective basic standards of protection against the ionizing radiations and the security of radiation sources (SAFETY SERIES In the 115), any exposure from natural sources should be excluded from regulation. The value used as an exclusion limit is, in accordance with the guide of security of the International Agency of Atomic Energy (SAFETY STANDARDS SERIES No. RS-G-1.7), 1 Bq/g for the series of ^{238}U and ^{232}Th, being the cited value to the precursor ("father") of each series and 4 Bq/g for ^{40}K. This value was determined based on the global distribution of the concentrations of activity of these radioisotopes in the earth's crust.

If this reference value is used, it would not be necessary to take corrective action to reduce exposures.. Therefore, according to the references cited above I contend that commodities with ^{238}U and ^{232}Th with lower concentrations than the 1 Bq/g standard not be subject to the regulatory control for the purposes of radioprotection. Under these conditions, doses are not anticipated that exceed the limit of exemption of 0,3 $mSvy^{-}1$. For activities that exceed the reference (e.g up to 10 times bigger) the regulatory authority will need to do a controlled evaluation on a case-by-case basis. The severity of the applied measures depending upon the level of the risk associated with the material.

As for radon, in accordance with the Safety Series 115, action should be taken when the levels of annual average concentration will be between 200 and 600 Bq/m^3 above 4.0 pCi/L in accordance with the guidance of US EPA. Actions of remediation are only justifiable at greater than 600 Bq/m^3 or 20 pCi/L range where cost-benefit analyses should be considered,.

Objective

This work aims at to the evaluation of the rate of emission of radon gas from Brazilian ornamental rocks, in view of the health of the human beings and the influence of these on the economy of the country.

Materials and Methods

In this work, three types of granite samples known commercially as *Crema Bordeaux*, *Mombassa* and *Golden*, were analyzed. These samples were selected because of the high tax of exportation for use in international the civil construction For the evaluation, samples, in the form of non-resin, polished plates (15 cm x 30 cm x 2 cm) were selected.

3.1. Rocks in the Natural State

For this study, sample preparation was not necessary. The testing apparatus consisted of a 20 L steel chamber and stamped removable covers for retention of the gas. Previously, the covers had been tested in order that confirm no leakage. Two plates of ornamental rocks, with known radiological potential were left in the box for eight days. To aid in mixing, a micro fan was used. Radon from the box was injected into a gas radon analyzer (*Alpha Guard 2000 PRQ Genitron Instruments*) in cycles of one and two hours. The in-growth of radon in the chamber was allowed to continue for a period of 7 half-lives. .

It is possible to know the tax of gas emission on each sample of the chamber by means of equation 1.

$$A_t = A_0 (1- e^{-\lambda t}) \quad (1)$$

where λ is the constant of nuclide decline on question and A_0 is the final value of the activity during 7 t ~ $T_{1/2}$, approximately 27 days in the case of the gas Rn. The unit of

the final activity A_0 is Bq m^{-3}. This value multiplied for the constant of decline of the radon (λ = 2,724 x 10^1 s^{-1}) and for the reason enters the volume of the container (V = 20,0 x 10^{-3} m^3) and the area of the granite, F, allows to get the rate of exhalation of radon for unit of area of this, that is defined as the flow of set free radon of the surface of the analyzed material, and, in Bq m^{-2} s^{-1}, that is, Becquerel per square meter per second, represented in equation 2.

$$E = A_0 \lambda (V / F) \quad (2)$$

In view of the comparison of the gas radon emission with the amount of radium in the samples the "final rate of emission of Rn for unit of mass" with unit in Bq kg^{-1} was calculated, using equation 3.

$$C_{Rn} = (A_0 V) / m \quad (3)$$

where m is the mass of the sample and A_0 and V already had been defined in the Eqs. (1) and (2), respectively.

3.2. Worn out rocks

For spectral gamma analysis, the samples were crushed, milled, screened and dried. For this analysis, the plates of the samples were crushed and then taken for milling. Dust from the milling process was controlled with aid of a sprayer (Fritsch). During the spraying, caution was used so that a majority of the samples would be between 0.177 mm and 0.149 mm after the screening. This size was considered optimal for gamma spectral analysis. Small amounts of sample in the container of the spray had been detritus of fish (titanium covered to prevent any type of contamination of the samples), and centrifuged at 400 r.p.m. for 2 minutes. After removal, the displaced samples were placed in a stack of bolters of 2,360 mm to remove the titanium balls of the spray, 0.177 mm and 0.149 mm, respectively. The samples were then put in a Ro-tap agitation device, for about 10 minutes. This allowed for total separation of the sample in the bolters. Although preference was given for samples between 0.177 mm and 0.149 mm, samples less than 0.149 samples were also used. Before mixing the samples were homogenized and the grain distribution was determined (e.g. what percentage was above or below 0,149 percentage mm). After the screening, the samples were ready for spectra-gamma analysis to allow for secular equilibrium, the samples were allowed placed in sealed containers for 30-days. A high efficiency gamma detector (HPGe Germanium) was used to measure the gamma emissions of ^{40}K and the daughters of the series of U and Th. Counting time varied from 8 to 16 hrs depending on the activity (Bq/g) of the material. The results were then compared to the threshold values listed by the International Agency of Atomic Energy (AIEA).

Results and Discussions

4.1. Granulometric Analysis

Table 1 indicates the results of the grain sized analysis of the *Crema Bordeaux, Mombassa* and *Golden samples.*

Table 1: analysis of the Grain size in which Sample

Granulometric Fraction (mm)	*Crema Bordeaux* Mass (%)	*Mombassa* Mass (%)	*Golden* Mass (%)
+ 0,177	0	0	0
- 0,177 + 0,149	10,29	16,52	10,59
-0,149	89,71	83,48	89,41
Total	100	100	100

4.2. Gas Emission ^{222}Rn

Figure 1 represents the results of the test of evaluation of the rate of gas radon emission in the samples in its natural state, being able itself to observe that the rates for the samples *Crema Bordeaux, Mombassa* and *Golden* had been very low, 3,10 x 10^{-3} ($Bq/m^2/s$), 1,60 x 10^{-3} ($Bq/m^2/s$), 6,84 x 10^{-4} ($Bq/m^2/s$), respectively.

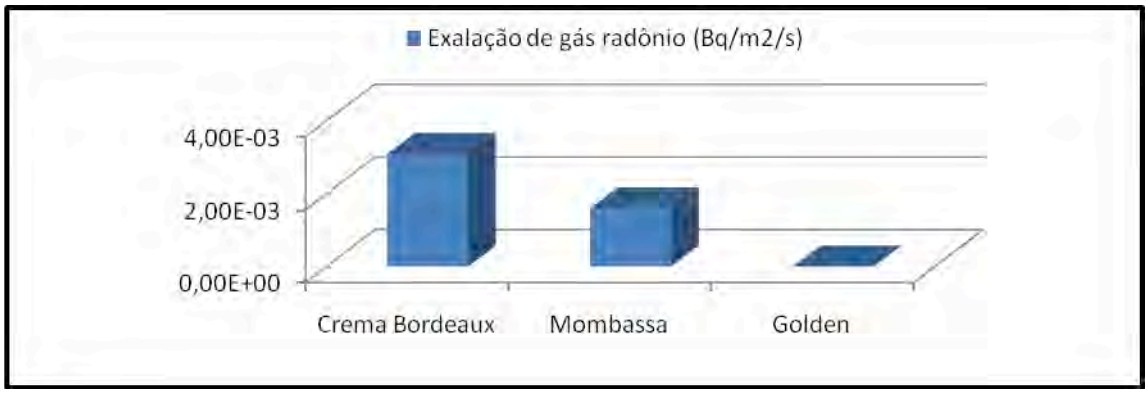

Figure 1: Rate of radon's emission in each sample in its natural state

4.3. Radiometric analysis / Radiochemical

Figure 2 Radionuclide´s Distribution in the Samples

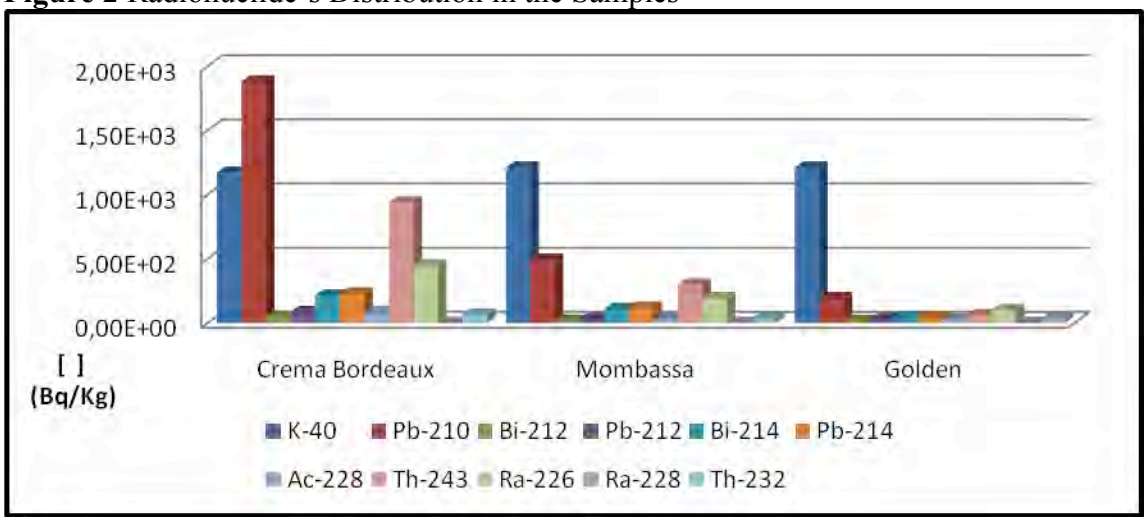

Figure 3 Main Radionuclides

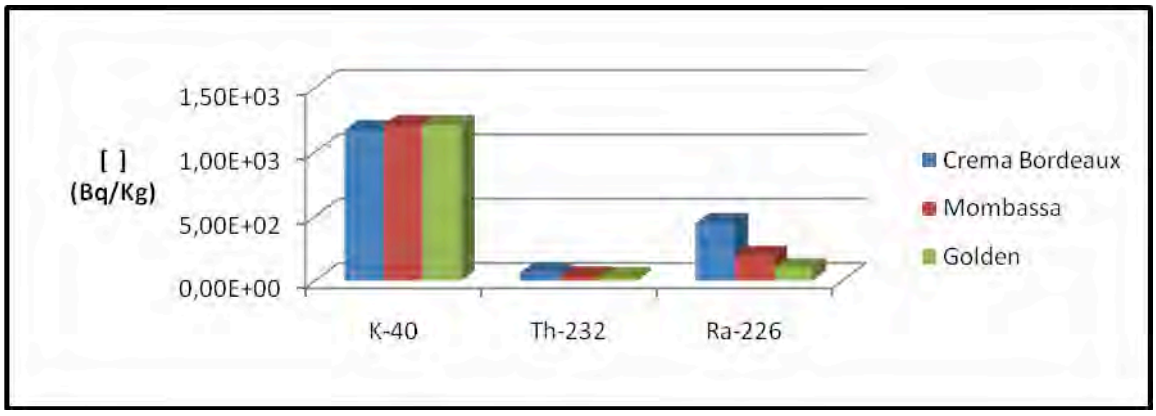

Figure 2 shows the results of the analyses of the three samples. The determination of Frog, K, Th and gamma had been carried out after the 30 days and with a detector of germanium, as said previously in the methodology. The determination had been made by counting alpha and beta, in a proportional detector of low background, after chemical separations and the measures of Thorium for spectrometry with *arsenazo III*.

Figure 3 shows the primary radionuclides measured in the samples.

Conclusion

Compared to international standards, the findings of this study indicate that radon emanation from these samples is very small. Therefore exposure risk from these types of granites would be minimal.

References

Anjos, R. M., Veiga, R. T. Soares, A.M.A. Santos, J.G. Aguiar, M.H.B.O., Frascá, J.A.P. *et al*. Natural Radionuclide Distribution in Brazilian Commercial Granites, Radiation Measurements, 2004.

Brodsky, A. Handbook of Radiation Measurement and Protection. CRP Press, available in: http://www.physics.isu/radiumf/natural.html, 1978.

Chyi, L. L. Radon Testing of Various Countertop Materials Final Report, 2008.

Craig Freudenrich, Ph.D., Marshall Brain. Available in: http://ciencia.hsw.uol.com.br/radonio1.htm. Access on: December 12th, 2008.

Fernandes, H.M.; Rio, M. A. P.; Franklin, M.R. Impactos Radiológicos da Indústria do Fosfato. Série Estudos & Documentos, n. 56, ISSN 0103-6319, CETEM, 2004.

HyperPhysics. Available in: http://hyperphysics.phy-astr.gu.edu/hbase/nuclear/radon.html. Access on: January 20th, 2009.

IARC - Agência Internacional de Pesquisa em Câncer.

Office of the Surgeon General – OSG. Available in: http://www.surgeongeneral.gov/. Access on: February 25th, 2009.

Rosa, R.. Exposição Potencial a Radiação Natural no Interior de Residências Devido ao Uso do Fosfogesso na Indústria da Construção Civil. [Tese de Mestrado]. Instituto de Biofísica Carlos Chagas Filho, Universidade de Federal do Rio de Janeiro. 122p., 1997.

US EPA - United States Environmental Protection Agency. Available in: http://www.epa.gov/. Access on: January 21th, 2009.

Whitfiel J. M.; Rogers, J. J. W.; Adams J. A. S..The relationship between the petrology and the thorium and uranium contents of some granitic rocks. Geochimica et Cosmochimica Acta 1959:17: 248-271.