

AN EVALUATION OF THE SCIENTIFIC PRODUCTION ON THE PRESENCE OF RADON IN BRAZILIAN ORNAMENTAL ROCKS

UMA AVALIAÇÃO DA PRODUÇÃO CIENTÍFICA SOBRE A PRESENÇA DE RADÔNIO EM ROCHAS ORNAMENTAIS BRASILEIRAS

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RESUMO - Em 2008, foi mencionado que as rochas ornamentais brasileiras poderiam conter altas concentrações de radônio. A questão tornou-se motivo de mobilização entre diversas instituições, incluindo a Comissão Nacional de Energia Nuclear (CNEN), tendo em vista que o Brasil é um grande exportador de rochas ornamentais. O motivo dessa preocupação é porque o radônio é um gás radioativo que emana de rochas e solos e tende a se concentrar em espaços fechados como minas subterrâneas, estacionamentos subterrâneos e habitações, sendo, portanto, considerado um dos principais contribuintes para a dose de radiação ionizante recebida pela população em geral. Nesse sentido, foi realizada uma revisão bibliográfica com o objetivo de gerenciar o conhecimento científico sobre a presença de radônio em rochas ornamentais brasileiras no Banco de Teses e Dissertações (BTD) da Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES) a fim de verificar a produção na área. Foram encontrados 178 estudos sobre radônio em geral, dos quais 14 estudos (7,87%) eram sobre a presença de radônio em rochas ornamentais brasileiras. O objetivo desta busca foi obter mais informações sobre as pesquisas sobre radônio em rochas ornamentais brasileiras, e em especial identificar estudos sobre uma maior variedade de rochas ornamentais produzidas no Brasil. Essa importância se deve principalmente ao fato de que uma grande variedade de rochas ornamentais brasileiras está presente na indústria da construção civil, utilizadas para revestimentos de pisos e fachadas. Portanto, pode-se concluir que a gestão científica do conhecimento sobre o radônio emanado das rochas ornamentais brasileiras em sua ampla variedade é de extremo interesse científico e econômico.

Palavras-chave: Radônio. Rochas ornamentais. CNEN. CAPES. Gestão do Conhecimento Científico. Brasil.

ABSTRACT - In 2008, it was mentioned that Brazilian ornamental rocks could contain high concentrations of radon. The issue became a reason for mobilization among several institutions, including the National Nuclear Energy Commission (CNEN), given that Brazil is a major exporter of ornamental rocks. The reason for this concern is because radon is a radioactive gas that emanates from rocks and soils and tends to be concentrated in closed spaces such as underground mines, underground parking lots and dwellings, and is therefore considered one of the main contributors to the dose of ionizing radiation received by the general population. In this sense, a bibliographic review was carried out with the aim of scientific knowledge managing about the presence of radon in Brazilian ornamental rocks in the Theses and Dissertations Database (BTD) of the Higher Education Personnel Coordination (CAPES) in order to verify production in the area. A total of 178 studies on radon in general were found, of which 14 studies (7.87%) were on the presence of radon in Brazilian ornamental rocks. The aim of this search was to obtain more information about research on radon in Brazilian ornamental rocks, and in particular to identify studies on a greater variety of ornamental rocks produced in Brazil. This importance is mainly due to the fact that a wide variety of Brazilian ornamental rocks are present in the construction industry, used for flooring and facade coverings. Therefore, it can be concluded that the scientific management of knowledge about radon emanating from Brazilian ornamental rocks in their wide variety is of extreme scientific and economic interest.

Keywords: Radon. Ornamental rocks. CNEN. CAPES. Scientific Knowledge Management. Brazil.

INTRODUCTION

The issue of Brazilian ornamental rocks and radioactivity

In 2008, in the United States, the fact that Brazilian ornamental rocks could contain high concentrations of radon and also high rates of gamma exposure was mentioned. This concern has become so expressive that several countries in Europe and Asia have imposed barriers to the commercialization of Brazilian granites. These

restrictions ended up making the Brazilian ornamental rocks market worried to the point that representative organizations of the sector sought the Nuclear Energy Commission (CNEN) and its institutes, such as the Institute of Radiation Protection and Dosimetry (IRD), requesting assistance regarding the assessment of levels radiation, since ornamental rocks are rarely evaluated for radiation in Brazil (Gavioli et al.,

2009; Moura et al., 2011).

This is because the industrial sector of Brazilian ornamental rocks is still a major exporter, in terms of value and physical volume, to several countries; it produces a wide variety of granites, marbles, quartzites, etc., reaching a total of about 500 different types of rocks. Such undue exposures would supposedly be caused by the exhalation of radon from “exotic” granites that would be increasing the risk of lung cancer. The reason for this concern is because radon is a major contributor to the dose of ionizing radiation received by the general population, hence the need to identify and quantify the levels of radioactivity and the various contributions in different building materials (Gavioli et al., 2009; Moura et al., 2011).

The study of radionuclide concentrations in granitic rocks is important for two reasons: technical (since these concentrations can be used for the petrographic classification of granites) and radiological (since the presence of radionuclides provides an increase in the levels of radiation present in civil construction). In the areas of geology and environmental radiological protection, granites are important examples of rocks that present in their constitution a natural enrichment of radionuclides - mainly uranium-238 (^{238}U), thorium-232 (^{232}Th) and potassium-40 (^{40}K) - in such a way that the concentrations of these radionuclides are correlated with their respective mineral compositions and general petrological characteristics (Gavioli et al., 2009; Moura et al., 2011).

It can be said that the gamma radiation existing inside a residence, coming from building materials, is mainly caused by the decay of radionuclides belonging to the ^{238}U , ^{232}Th and ^{40}K chains. In the ^{238}U and ^{232}Th chains there are a total of 16 gamma radiation emitters that emit radiation with an emission rate greater than 10^{-3} photons per decay (due to the fact that this gamma radiation is generated isotropically, only a fraction of the emissions originals is likely to reach the exposed individual inside the building) (UNSCEAR, 2000).

Soil gas infiltration is recognized as the most important source of residential radon. Other sources, which include building materials and water taken from wells, are of minor importance in most circumstances. In most buildings, the air pressure at ground level is slightly lower than outside because the air inside the ground is

warmer. This generates a passage of air from the ground to the interior of the building and, together with this air, ^{222}Rn will also be transported. The main access routes are spaces between floors and walls, floor cracks and voids around pipes and cables. There are seasonal variations in indoor ^{222}Rn levels, corresponding to variations in the average outdoor temperature (^{222}Rn levels in winter are generally higher than in summer) (UNSCEAR, 2000; EPA, 2004).

Radon is a naturally occurring radioactive gas found almost everywhere on the Earth's crust. It emanates from rocks and soil and tends to concentrate in enclosed spaces, such as underground mines, underground parking lots and homes, as there is less space for ventilation. Therefore, the ingestion of water or the inhalation of air with high levels of radon gas can expose sensitive cells of the respiratory and gastrointestinal tracts to ionizing radiation, which represents a risk to the health of the population. In the United States, for example, radon is believed to be an important cause of lung cancer, killing about 10,000 Americans each year, due to illness due to the occurrence of some types of cancer (UNSCEAR, 2000; EPA, 2004).

Considering that people spend more than 80% of their time indoors, the duration of exposure to internal and external radiation from building materials is extended. Therefore, the level of external and internal exposure depends on the concentration of radionuclides in the building materials and the duration of stay in the indoor environment. Most indoor exposure to radon occurs through the emission of alpha particles, although beta particles and gamma radiation are also produced. There is a general consensus among scientists about the risks of lung cancer related to alpha radiation, which comes from radon. The concentrations of this gas in the environments depend on the characteristics of the residences, such as ventilation system, types of building materials, among others, more significantly depending on the seasons of the year (UNSCEAR, 2000; EPA, 2004).

There is variation in radon concentration throughout the day and also at certain times of the year, with the highest concentrations occurring in the early hours of the day and the lowest in the late afternoon. However, the highest radon values are found in the autumn or winter months and have minimums in the spring. Meteorological factors (rain, wind, temperature,

pressure, among others) influence the rate of exhalation or emanation of radon in the soil and its dispersion in the atmosphere, causing the rate of exhalation from the soil to increase during periods when atmospheric pressure decreases. Concentrations decrease the higher and further away the measurement is from the ground. The highest radon concentrations occur in the most arid regions, since cracked soils tend to increase the transport of ^{222}Rn (UNSCEAR, 2000; EPA, 2004).

Radon

The radon is a radioactive, natural gas, present virtually everywhere in the earth's crust, which emanates from rocks and soils and tends to be concentrated in closed spaces such as underground mines, underground parking lots and dwellings, as there is less space for ventilation. Radon is a colorless, odorless, tasteless gas, in addition to being 7.58 times heavier than air and over a hundred times heavier than natural hydrogen, it originates from radium, a member of the uranium decay series and thorium (UNSCEAR, 2000).

^{222}Rn is a radioactive gas with alpha emission and decays with a half-life of 3.82 days, emitting alpha particles with an energy of 5.49 MeV. The last element to decay is ^{210}Pb , which, although not radioactive, lead can bind to bones, causing contamination by heavy metals. After being absorbed, it remains in the blood, with a half-life of 1 to 2 months, and can then be excreted or distributed in soft tissues. Over time, it is deposited in bones, teeth and hair, which in turn can cause serious damage to health, and can even cause death. There is no major concern with ^{220}Rn because it has a very short half-life (UNSCEAR, 2000; EPA, 2004).

The radon-222 (^{222}Rn) isotope is an alpha emitter ($T_{1/2} = 3.82$ days) and, together with its non-gaseous children polonium-218 (^{218}Po) and polonium-214 (^{214}Po), is responsible for approximately 50% of the equivalent effective dose produced by natural ionizing radiation. There is also radon-219 (^{219}Rn) and radon-220 (^{220}Rn) which are products of the ^{235}U and ^{232}Th decay series, respectively. They have a very short half-life when compared to ^{222}Rn , being that of ^{219}Rn of 3.96 seconds and that of ^{220}Rn of 55.6 seconds, therefore, ^{222}Rn is the only one capable of migrating into homes and raising concerns in the health area (UNSCEAR, 2000; EPA, 2004).

Radon is a chemically inert, colorless, odorless

and tasteless gas. As a gas, it can spread into human environments through construction materials, soil and water, exposing individuals to radiation. It is 7.58 times heavier than air and more than a hundred times heavier than natural hydrogen (EPA, 2004).

It is generated in all materials in general and is more significant in those with large amounts of uranium, thorium and radium. The higher the concentration of these elements, the higher the concentration of radon. They originate from the radium isotopes ^{226}Ra for ^{222}Rn and ^{224}Ra for ^{220}Rn , and the γ -emitting decay products of ^{222}Rn are ^{214}Pb and ^{214}Bi , which are found in radioactive secular equilibrium with ^{226}Ra only if sealed to prevent radon from escaping, while the decay products of ^{220}Rn are ^{212}Pb , ^{228}Ac , ^{208}Tl and ^{212}Bi , which are found in radioactive secular equilibrium with ^{224}Ra only if sealed to prevent ^{220}Rn from escaping. From these decay products, the estimated activity concentration in Bq.kg⁻¹ of ^{238}U and ^{232}Th can be obtained (Gavioli et al., 2009; Moura et al., 2011; UNSCEAR, 2000; EPA, 2004).

Disturbance of the radioactive equilibrium in building material due to radon emanation can result in ^{214}Pb and ^{214}Bi activity levels in the material being lower than that of ^{226}Ra . Radon transport is mainly due to diffusion and forced flow. When radon is inhaled, most of the radon is exhaled, but its daughter products remain in the inner walls and membranes of our respiratory system and continue to cause ongoing damage due to their alpha decays. The dose of alpha particles is delivered directly to the bronchial tissue, creating a potential for cardiogenic lung cancer and in some cases kidney disease, in addition to the risk of causing DNA mutation (Gavioli et al., 2009; Moura et al., 2011; UNSCEAR, 2000; EPA, 2004).

In this sense, this work was developed in order to carry out a bibliographic review aimed at Scientific Knowledge Management in the area of study on the presence of radon in Brazilian ornamental rocks. This is because it is understood that the understanding of scientific knowledge itself, by a teaching and research institution, ends up promoting advances in its activities, helping in the technological innovation of its respective area of activity (Amorim et al., 2021). The main objective is to evaluate research that focuses on the identification and quantification of public exposure to radon gas in Brazilian ornamental rocks.

MATERIALS AND METHODS

This paper reports a qualitative-quantitative investigation (Gil, 2007), through the search using the keyword “radon” in the Database of Theses and Dissertations” (BTD) (BRASIL, 2022) of the Higher Education Personnel Coordination (CAPES) in order to check production in the area.

Works were selected whose keyword was found either in the title, or in the abstract or in the keywords, or even in the body of the work [Amorim et al., 2021; Gil, 2007]. An analysis will then be made of master's dissertations and doctoral theses in the specific search on Brazilian ornamental rocks.

RESULTS AND DISCUSSION

A total of 178 works were found on radon in general, 54 theses and 124 dissertations since 1996 (it is important to highlight that there are other studies carried out in Brazilian institutions focusing on radon before 1996, but unfortunately, they are not included in the database used in the article).

Of this total, fourteen works (three dissertations and eleven theses) were related to Brazilian ornamental rocks, which correspond to about 7.87% of the total on radon. The production on the presence of radon in Brazilian ornamental rocks is restricted between the years 2005 and 2017. Works related to construction

materials were disregarded.

These works were defended in the years of:

- 2005 - one work (one thesis);
- 2011 – two works (one dissertation and one thesis);
- 2012 – one work (one thesis);
- 2013 – three works (three theses);
- 2014 – two works (two dissertations);
- 2016 – three works (three theses); and
- 2017 – two works (two theses).

Thus, a graph can be created showing the temporal evolution of production in the area (figure 1).

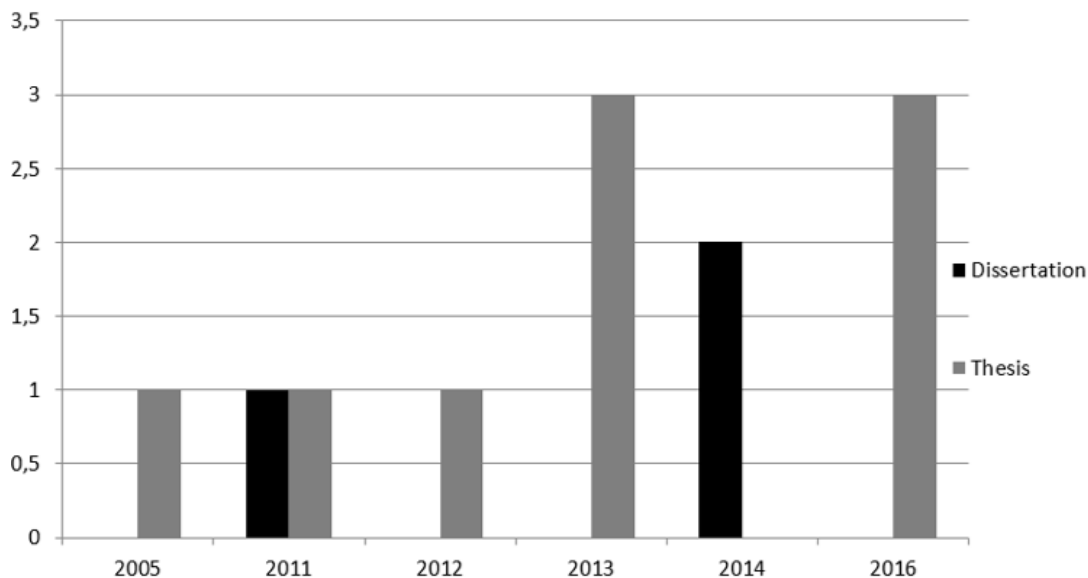


Figure 1 - Temporal evolution of the production of works on the presence of radon in Brazilian ornamental rocks.

The works were found in four Knowledge Areas (Exacts and Earth Sciences - eight works; Engineering - four works; Multidisciplinary - one work; Social and Applied Sciences - one work) and in five Assessment Areas (Geosciences – six works in Geology; two works in Regional Geology; Engineering II – one work in Nuclear Engineering and one work in Mineral Engineering; Interdisciplinary – one work; Architecture, Urbanism and Development – one

work; Engineering IV - one work in Biomedical Engineering) from CAPES. Works were found in five Teaching Institutions (IEs) and in seven Postgraduate Programs. The Programs were in (figure 2):

- Architecture and Urbanism (one work – one thesis);
- Electrical Engineering and Industrial Informatics (one work – one thesis);
- Mineral Engineering (one work – one thesis);

- Geology (six works – two masters and four theses);
- Nuclear Technology (two works – two theses); and
- Regional Geology (two works – two theses);
- Radiation Protection and Dosimetry (one work – one dissertation).

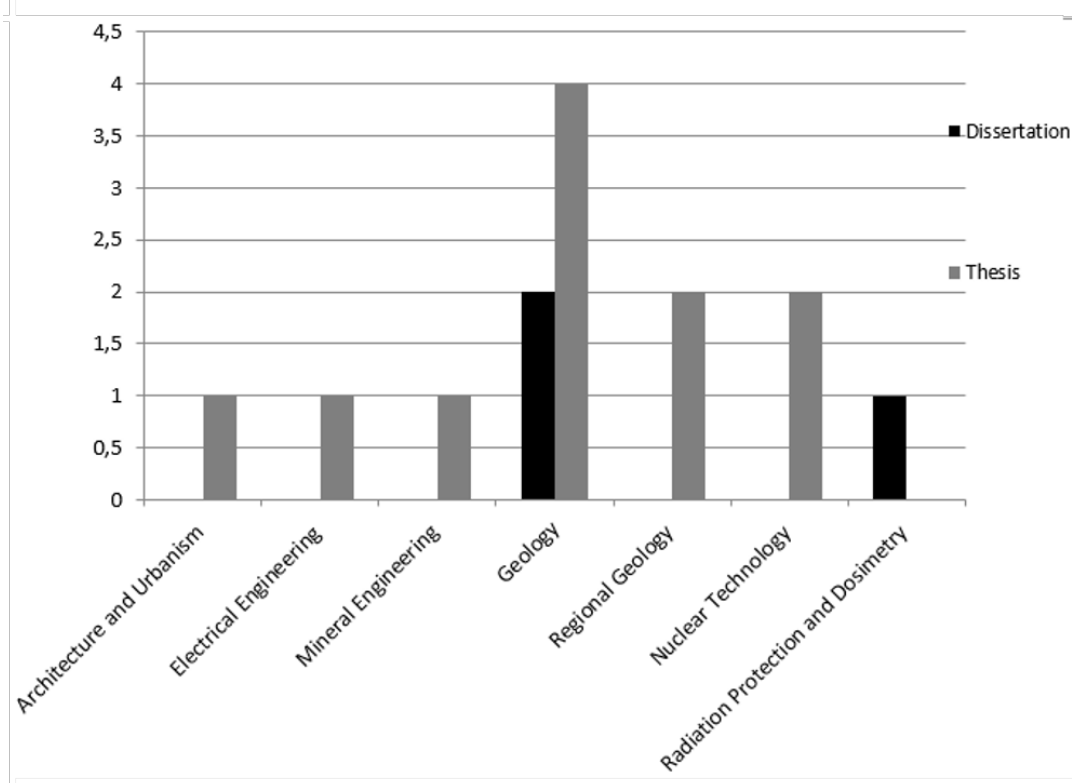


Figure 2 - Programs that developed work on the presence of radon in Brazilian ornamental rocks.

The works belong to the following IEs (Figure 3):

- Institute of Radiation Protection and Dosimetry (IRD) – one work (one dissertation);
- University of São Paulo (USP) – four works (four theses);
- São Paulo State University Júlio de Mesquita Filho/Rio Claro (UNESP) – three works (one dissertation and two theses);
- Federal University of Ceará (UFC) – five works (one dissertation and four theses); and
- Federal Technological University of Paraná (UFTPR) – one work (one thesis).

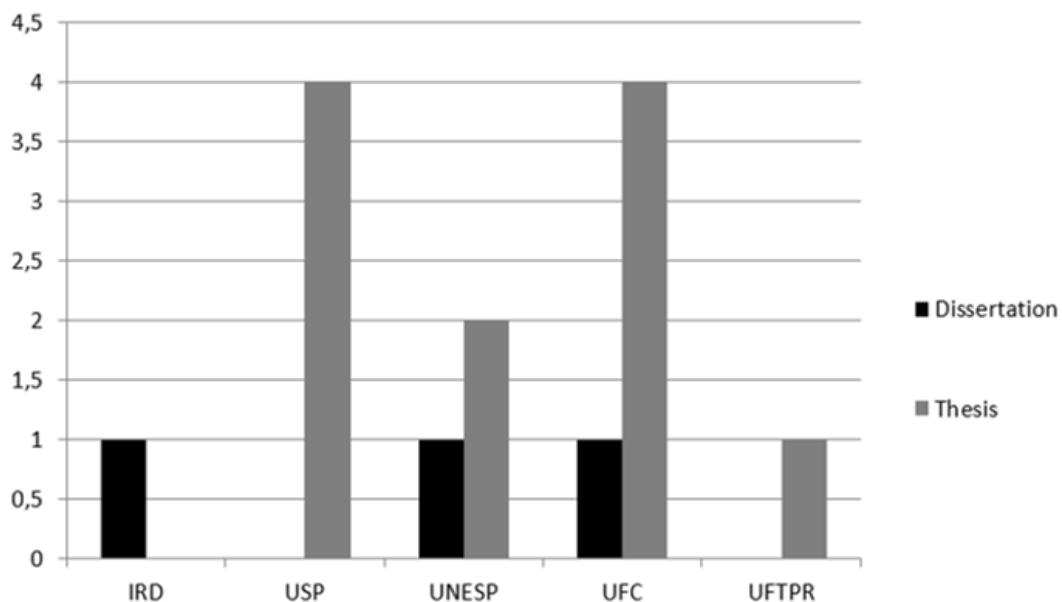


Figure 3 - IEs that developed works on the presence of radon in Brazilian ornamental rocks

groups in activity, as well as their areas of concentration, in order to carry out a more detailed assessment of research in general and in the nuclear area in the identification of coordinated works with specific objectives in meeting the international recommendations.

Thus, it is concluded that the Management of Scientific Knowledge on Radon emanating from Brazilian ornamental rocks is extremely important in helping new investigations and the construction of knowledge on the subject in the future.

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APPENDIX

Results reported in the literature on studies evaluating the rate of Radon in Brazilian Ornamental Rocks.

Work/ Keywords/Abstract	METHODS	RESULTS/ CONCLUSION
<p>1) MOURA, C.L.M. Natural radioactivity and emanation of Radon-222 in ornamentais rocks from different magmatic series used as internal coating. Ph. D. in Regional Geology, São Paulo State University Júlio de Mesquita Filho, 2005. Keywords: Natural Radioactivity; Dimension Stones; Granites; Gamma-ray Spectroscopy. Abstract. This paper reports the natural radioactivity of Brazilian igneous rocks that are used as dimension stones, following the trend of other studies on the evaluation of the risks to the human health caused by the rocks radioactivity as a consequence of their use as cover indoors.</p>	<p>Gamma-ray spectrometry has been utilized to determine the ^{40}K, ^{226}Ra and ^{232}Th activity concentrations in 14 rock types collected at different quarries.</p>	<p>The following activity concentration range was found: 12.18–251.90 Bq/kg for ^{226}Ra, 9.55–347.47 Bq/kg for ^{232}Th and 407.5–1615.0 Bq/kg for ^{40}K. Such data were used to estimate R_{aeq}, H_{ex} and I_{g}, which were compared with the threshold limit values recommended in literature. They have been exceeded for R_{aeq} and H_{ex} in five samples, where the highest indices corresponded to a rock that suffered a process of ductile–brittle deformation that caused it a micro-brecciated shape. The exhalation rate of Rn and daughters has also been determined in slabs consisting of rock pieces 10 cm-long, 5 cm-wide and 3 cm-thick. It ranged from 0.24 to 3.93 Bq/m²/h and exhibited significant correlation with eU ($1/4^{226}\text{Ra}$), as expected. The results indicated that most of the studied rocks did not present risk to human health and may be used indoors, even with low ventilation. On the other hand, igneous rocks that yielded indices above the threshold limit values recommended in literature may be used outdoors without any restriction or indoors with ample ventilation.</p>
<p>2) AMARAL, P.G.Q. Radiometric and Radon Exhalation Characterization in Processed Silicatic Ornamental Rocks in the State of Espírito Santo. Master in Geology, São Paulo State University Júlio de Mesquita Filho / Rio Claro (UNESP), 2011 Keywords: Dimension Stone; Radon Gas; Radioactivity. Abstract. This work carried out assessments on the radiometric behavior and exhalation of radon gas in dimensional and surfacing stones exploited in the Brazilian states of Minas Gerais (MG) and Espírito Santo (ES). The rocks usually contain different levels of the radionuclides ^{87}Rb, ^{40}K, ^{238}U, ^{235}U and ^{232}Th, the last three forming radioactive decay series, where different Rn isotopes occur. Despite its low natural activity concentration in the environment, ^{222}Rn (half life = 3.8 days) is an important radioactive noble gas because can generate the heavy metals ^{218}Po, ^{214}Pb, ^{214}Bi and ^{214}Po which can settle in the lungs, causing major diseases in the respiratory tract, including the induction of metastasis.</p>	<p>Ten silicate rocks of variable composition and commercial use have been selected for evaluating the radon presence in human environments. They have been benefited by companies in Cachoeiro de Itapemirim (ES) and include magmatic (diorite, syenite, charnockite, monzogranite, granitic pegmatite) and metamorphic rocks. The study involved the acquisition of U, Th and K radiometric data, as well the monitoring of the ^{222}Rn gas exhalation, coupled with petrographic and physical rock indices.</p>	<p>The U and Th content range was 2.9–37 and 0.3–84ppm, respectively, where the radioelements content was consistent with the presence or absence of accessory minerals hosting them. The amount of radon emanated from the rocks was directly related to the petrography of each material, especially the degree and types of microcracks and the contacts between minerals that are determinants of the rocks microporous net, thus, leading to a greater or lesser permeability, which in turn controls the exhalation. The comparison of the ^{222}Rn generated by the rock with the amount effectively exhaled indicated that the exhalation rate is negligible and also no correspondence between the more Rn-productive and Rn-exhalative rocks. Simulations of the application of the studied materials as flooring indoors have been performed in order to evaluate the cumulative radiation levels generated by the radon gas exhalation of the rocks. The results indicated that nine samples yielded values below the 4pCi/L guideline value established by EPA, whereas, the pegmatite provided 7.92pCi/L that is above it.</p>
<p>3) SILVA, F.D.O. Technological and radiometric characterization of a diabase in the region of Apuiarés-CE. Ph. D. in Geology, Federal University of Ceará (UFC), 2011. Keywords: Radon; Dimension stones; Porphyritic olivine diabase.</p>	<p>The radiometric measurements were performed using the methods of gamma spectrometry and detection of active exhalation of radon gas to monitor the amount of ^{222}Rn. Added to the main objectives of this study were also made petrographic directed to the recognition of the mineralogical composition and textural features among other features.</p>	<p>The physical indices showed higher than average for siliciclastic rocks in Brazil, namely, apparent specific gravity, porosity and water absorption with respectively 3113 kg / m³, 0.17% and 0.06%. The values of uniaxial compressive strength also exceed the average for Brazilian siliciclastic rocks and also within the limits of ASTM, with an excellent compressive strength of 192 MPa.</p>

<p>Abstract. This paper presents the technological and mineralogical characteristics of a diabase, which outcrop in the form of dike is located in the district Canafistula in Apuiarés - CE. In order to assess their qualities for application as ornamental, were carried out technological tests of fitness levels and physical-mechanical, based on standards from ABNT, as well as radiometric measurements of the amount of uranium, thorium and radioactive potassium in the rock, beyond radon exhalation rate.</p>	<p>The petrographic analysis allowed to classify the rock as olivine diabase, porphyritic texture, composed of mineralogically titanite + olivine + plagioclase + apatite ± opaque ± iddingsite clorofeita ± biotite. This has lithotype ranging gray to black, with isotropic structure. The degree of microcracking of the rock is located around 3.53 / mm², where 97.9% of the cracks are of the type intragrain and 2.1% are of the intergrain.</p>	<p>Regarding the 3-point bending test, the rock exhibited value of 20.4 MPa, well above the average for Brazilian siliciclastic rocks and ASTM. The behavior of the samples when subjected to wear AMSLER, gave a performance with 0.66 mm wear for a journey of 1000m below the average for Brazilian siliciclastic rocks. The analysis by gamma spectrometry provided values for 238U and 40K, of 0.54 ppm and 2.88%, and when transformed into Bq.kg⁻¹, corresponded to 6.8 and 48.3, respectively. The final activity of radon emanation (CRN) and radon exhalation rate (E) registered a content of <0.016 Bq.Kg⁻¹ (<0.432 pCi.kg⁻¹) and <0.002 Bq.m-2h⁻¹ (<0.054 pCi.m-2.h⁻¹). Within this context, the olivine diabase porphyry shown to have diverse applications such as ornamental material for optimal mechanical characteristics and low concentrations of radioactive elements as well as low emanation and exhalation of ²²²Rn, that due to its material characteristics of basic composition, good mechanical strength, low porosity and low amount of microcracking.</p>
<p>4) SALES, F.A.C.B. Comparative study of technological characterization parameters in ornamental rocks and resin and non-resin coatings. Doctorate in Geology. Federal University of Ceará (UFC), 2012. Keywords: Ornamental and coatings; Resin; Technological characterization. Abstract. For intermediate analysis and technological characterization tests conducted on samples of resinated and non-resinated rocks with distinct structural and geological characteristics, we sought to verify the effectiveness of the resin with respect to physical and mechanical resistance of these rocks, as well as evaluating the reduction of agents that produce the same changes and pathology.</p>	<p>The investigation of this research has focused on three types of rocks: the first refers to a commercially called White Granite Blizzard, which is an isotropic rock with some degree of fracture, equigranular texture and average grain size of the fine and the second was represented petrographically for a garnet-biotite-muscovite gneiss, commercially known as Casa Blanca, having a coarse-grained, with lineations and foliations with a high presence of micaceous minerals, and the third is a volcanic rock, trade name Wood Stone, material considered exotic because of their textural heterogeneity, with a high degree of fissuring, having very fine grain. After the results of tests and analysis, they were correlated and evaluated the effectiveness of the resin, being featured in that it produces significant improvements in physical and mechanical resistance of these rocks, especially regarding the levels of porosity and water absorption, which are the gateway of the causative agents of change and generations of pathologies in ornamental and coating.</p>	<p>Research has shown that materials with restricted use, or even inappropriate to certain environments, due to its physical and mechanical resistance, using the resin, the same could be used without restrictions. The importance of testing materials whose marketing is made using resin was evident due to changes often significant parameters of the results of characterization of technology, which serve as reference in the specification suitable for use as coating materials or ornamental, the construction works. The apparent porosity and water absorption were reduced by 9, 7% and 91.46% respectively after the material is resin.</p>
<p>5) AQUINO, R. R. Evaluation of natural radioactivity in commercial marbles and granites in the state of Espírito Santo. Ph. D. in Nuclear Technology, University of São Paulo (USP), 2013. Keywords: Granite; HPGe; Marble; Natural radioactivity; SSNTD. Abstract. In this work, the concentrations of natural radionuclides ²²⁶Ra, ²³²Th and ⁴⁰K in granite and marble samples were determined, considering the main extraction mining of Espírito Santo state, southeastern Brazil.</p>	<p>For all study sites, three samples were sealed in 100 ml high density polyethylene bottles. Each sample rested for 4 weeks to reach the secular equilibrium of ²³⁸U and ²³²Th series before measured by high resolution gamma spectrometry, and the acquired spectra were analyzed with the software WinnerGamma. The self-absorption correction was considered for all samples, using an expression and method specially developed for this purpose. The concentration of ²²⁶Ra was determined by the weighted arithmetic mean of the concentrations of</p>	<p>The activities determined by passive detection varied from 100 ± 10 Bqm⁻³ up to 2400 ± 300 Bqm⁻³, highlighting the biggest exhalation rates for granite Iberê Mombasa. Considering the marbles, activity values varied from 80 ± 10 Bqm⁻³ up to 200 ± 25 Bqm⁻³ highlighting only the Cintilante and Branco Extra with higher values. The values obtained for surface exhalation rate were approximately equal, except for granites Iberê Mombasa and Iberê Prado with values above 1 Bqm-2h⁻¹. The measures by gamma spectrometry showed that the ²²⁶Ra concentrations varied from 1.9±0.2 Bq.kg⁻¹ up to 483±55 Bq.kg⁻¹, with the highest value for granite Iberê Mombasa. The ²³²Th</p>

	<p>214Pb and 214Bi, the concentration of 232Th by the weighted arithmetic mean of the concentrations of 228Ac, 212Pb and 212Bi and the concentration of 40K by its single 1460 keV transition. The radium equivalent and gamma index were calculated from the activity concentrations of 226Ra, 232Th, and 40K. The emanated radon was measured using an exhalation chamber and the passive detector technique, with a solid state nuclear tracks detectors (SSNTD) being exposed in NRPB/SSI-H dosimeters. During exposure, a commercial detector CR-39® and a national plastic called Durolon® were used, the last was characterized for this purpose using a technique called double exposure and sensitivity intrinsic factor. The characterized plastic was efficient for the application and the calibration factor corresponded to 1.60 ± 0.10 trackscm²(kBqm³day)⁻¹ in relation to the CR-39 factor, equivalent to 2.8 ± 0.2 trackscm²(kBqm³day)⁻¹. The detector showed a higher efficiency at a higher dose.</p>	<p>concentrations ranged from 3.2 ± 0.4 Bq.kg⁻¹ to 224 ± 6 Bq.kg⁻¹, whose largest value was observed for the gray granite Corumbá. The ⁴⁰K concentrations ranged from 8.8 ± 1.8 Bq.kg⁻¹ up to 1642 ± 67 Bq.kg⁻¹, with the largest value observed for granite Iberê Vitará. The radio equivalent value showed that most samples can be defined as category I, below 370 Bq.kg⁻¹, except for the granites Cinza Corumbá, Iberê Crema Bordeaux and Iberê Mombasa that can be classified as class II (up to 740 Bq.kg⁻¹). The evaluated granites show internal and external exposure rates below 1.0 mSv.y⁻¹ except the granites Cinza Corumbá, Iberê Crema Bordeaux and Iberê Mombasa that exceed the value range of 1.0 to 3.2 mSv.y⁻¹ for this index. For the annual exposure dose, only the granites Gray Corumbá, Iberê Crema Bordeaux and Iberê Mombasa exceeded the 1.5 mSv.y⁻¹. For the alpha exposure index only the Iberê Crema Bordeaux and Iberê Mombasa granites indicate limitations when applying as surface material. However, considering the gamma exposure index, the granites Cinza Corumbá, Cinza Andorinha, Amarelo Icarai, Cinza Ocre, Iberê Crema Bordeaux and Iberê Mombasa have controlled application. In conclusion, the evaluated granites and all marbles evaluated have viable applications in different activity sectors and for different purposes and the granites that exceeded the proposed limits should not be applied in the interior of residences.</p>
<p>6) AZEVEDO, L.R.P. Radon Emanation in Ornamental and Cladding Stones in the State of Ceará, Brazil. Ph. D. in Regional Geology, São Paulo State University Júlio de Mesquita Filho/Rio Claro (UNESP), 2013. Keywords: Ornamental Rocks; Radon exhalation; Granites; Levels of U, Th and ⁴⁰K; Radioactivity. Abstract – This research aims to quantify the gas ²²²Rn exhalation and the corresponding levels of U, Th and ⁴⁰K in rocks used for ornamental purposes and coating in Ceará State. About 75% of the Ceará territory has been occupied by crystalline basement, providing favourable conditions for the occurrence of granite and other rocks with ornamental features such as marble, quartzite, and pegmatite, among others.</p>	<p>Considering the increasing use of rocks as coating material in interior environments, it had been selected and used fifteen different types of rocks exploited in Ceará State, including thirteen granites with different textural and structural aspects, a conglomerate and a pegmatite, which were submitted to petrographic analysis, determination of physical characteristics (apparent density, apparent porosity and water absorption), determination of the U level, Th and K by ICP-MS and monitoring the gas ²²²Rn amount emanated by these rocks. The rocks naturally present in their mineralogical composition determined concentrations of radioactive elements such as ²³²Th, ²³⁵U and ²³⁸U, which generate by radioactive decay the ²²⁰Rn, ²¹⁹Rn and ²²²Rn, respectively, being the latter one the aim of this research, once this element has been showing the longer half-life among the three radionuclides, about 3.85 days.</p>	<p>The U concentrations provided by the rocks had varied from 0.2 ppm to 13.6 ppm, which were used to determine the levels of ²²²Rn in Bq/kg generated by the rocks (which are between 2.47 and 167.82 Bq/kg) and then compared with the quantities of gas ²²²Rn effectively exhaled (between 0.09 and 6.04 Bq/kg). The results related to the U levels and the correspondent exhalation of the ²²²Rn gas, which had been provided by the rocks, had shown quite consistent with their petrographic characteristics and porosity of each material. The radon gas exhalations obtained in Bq/m³, for all the rocks studied in this work, were converted to pCi/L, showing values below the limit of 4 pCi/L recommended by the Environmental Protection Agency of the United States (EPA), varying from 0.04 pCi/L to 2.59 pCi/L.</p>
<p>7) FERREIRA, A.O. Evaluation of natural radioactivity in some granitic rocks in the State of Paraná and its use in civil construction. Ph. D. in Nuclear Technology, University of São Paulo (USP), 2013. Keywords: Dose; Exhalation; Natural Radioactivity; Radon. Abstract – Primordial, or terrestrial natural radionuclides, are found in different</p>	<p>Also, possible correlations between the ²²⁶Ra activity concentration, the ²²²Rn exhalation rate, density, porosity and chemical composition (oxide content) in these samples had been studied. The external dose was assessed by gamma-ray spectrometry with High-Purity Germanium detectors, where the activity concentration of the radionuclides ²³²Th, ²²⁶Ra and ⁴⁰K are the parameters used in</p>	<p>The results for this studies show that the annual effective dose ranged from (62 ± 3) μSv.y⁻¹ to (138 ± 1) μSv.y⁻¹ and the internal annual effective dose ranged from (0.39 ± 0.04) μSv.y⁻¹ to (70 ± 4) μSv.y⁻¹. These values are below the maximum limit of 1 mSv.y⁻¹ suggested by the European Commission of Radiological Protection, meaning that the granitic rocks evaluated can be used without radiological implications since the considered</p>

<p>amounts in the environment. In dwellings, an important dose increment is due to building materials, which contribute for both the external gamma dose from the radionuclides of the ^{238}U, ^{235}U and ^{232}Th series and the natural ^{40}K and the internal dose, due mainly to ^{222}Rn inhalation. Once granitic rocks are widely used both as construction materials or structural flooring, those rocks can become an important dose source, depending on the content of concentrations of radioactivity, and the construction application. In this work, a database for granitic rocks of the crystalline shield of Paraná (mainly in the Metropolitan Region of Curitiba, RMC), used in civil construction, was generated, evaluating in terms of radiological protection the external and internal dose increments, caused by the use of these materials.</p>	<p>dosimetric models (Dosimetric Indexes), which established limits in accordance with the form, amount and application of material of construction. For the calculation of the annual effective external dose it was assumed a room model with dimensions of 4 m x 5 m x 2.8 m and all walls internally covered with 2 cm thickness of granite and an annual exposure time of 7000 h as suggested by the European Commission of Radiological Protection for internal superficial coating materials. The internal exposure was assessed from the radon concentration in the air of the room model, simulated from the superficial exhalation rate of ^{222}Rn. The exhalation rate was determined by the passive detection technique with the Solid State Nuclear Track Detectors (CR-39) and the sealed can technique, assuming a ventilation rate of 0.5 h^{-1} and an annual exposure time of 7000 h.</p>	<p>scenario is obeyed. The values obtained for the contribution due to the internal dose ranged from 1 % to 78 % of the values obtained for the respective external dose showing the radon contribution varies strongly with the rock type. The results of the correlations between ^{222}Rn superficial exhalation rate, ^{226}Ra activity concentration, density, porosity and major oxides of the samples, showed that, in terms of influence in the emanation fraction of radon, the most important parameter is the density, due to low porosity and similarity in terms of chemical composition amid the studied samples.</p>
<p>8) LEAL, A.L.C. Evaluation of the dose associated with the use of granite in residential and commercial establishments. Master's in Radiation Protection and Dosimetry, Institute of Radiation Protection and Dosimetry (IRD), 2014. Keywords: Dosimetry. Radioactivity, Radon. Granite. Abstract – Many studies aims to get an estimate dose associated with the use of granite rocks such ornamental rocks and use as floor coating indoor, has increased in recent years. This interest is triggered due out of natural radionuclides levels found in accessory minerals that make up these rocks and due the radioactive decay of these radionuclides results in a radiological exposure indoor for the occupants.</p>	<p>The concentration values in 180 samples of Brazilian granitic rocks vary in a wide range: K-40 ranging from 190 to 2300 Bq.kg^{-1}, while Ra-228 (Th-232) varies from 1.9 to 530 Bq.kg^{-1} and R-226 between 4.9 to 600 Bq.kg^{-1}. The ratios between Ra-228 and Ra-226 vary from 0.1 to 43 indicating the strong weathering suffered by these rocks and about 40% of the samples had the activity gamma index, the internal risk index and the radio equivalent index greater than their reference limits. This could mean that the use of these rocks result in risk to health. However, the values of the estimated dose resulting from the use of granite is in residential and commercial establishment ranged from 0.02 mSv to 0.91 mSv and from 0.006 to 0.25 mSv, respectively. The largest contribution to the total dose is due external gamma exposure, then the thoron and radon.</p>	<p>Only two samples showed the highest external dose (0.3 mSv/y) and the highest inhalation dose due of radon was 0,008 mSv/y and was 0,17 mSv/y for thoron. These doses have very low values compared to the established dose level of 10 mSv/y. The concentrations of Rn-222 ranged between 0.09 and 11 Bq.m^{-3} and they are very below to the reference value of 300 Bq/m^3. Among the parameters that strongly influence the dose are the diffusion coefficient emanation rate, the emanation radon rate and, in menor degree, the renewal internal air rate. The simulations showed, in general, the results obtained for dose were below the limit recommended by the International Commission on Radiological Protection (ICRP) and is possible inferred that such reference index overestimate the doses and is necessary reviewed their formulations and used, since it has been so widely used. This work proposes a new index based on the approaches adopted by RESRAD BUILD.</p>
<p>9) NASCIMENTO, D.M. Technological characterization of an ultramylonite from the Marco-CE region. Master's in Geology, Federal University of Ceará (UFC), 2014. Keywords: Ultramylonite; Green Galaxy, Tucunduba; Technological characterization. Abstract: This dissertation presents the mineralogical and technological characteristics of the Granite Tucunduba, commercially known as Green Galaxy, located in the municipality of Marco - CE. Technological tests of physical indexes (bulk density, apparent porosity and water absorption apparent d'), uniaxial compression, bending in three and four points, AMSLER abrasive wear, impact and hard freezing and thawing body and speed of wave propagation were performed ultrasonic, based ABNT. To complement the data also quantified the rate of exhalation of radon.</p>	<p>Radiometric measurement was performed by applying the active detection method exhalation gas for monitoring the amount of ^{222}Rn. With the choice of the tests described, aimed to evaluate the quality of rock for use as ornamental stone. Was added to the main objectives of this work focused on the recognition of the mineralogical composition and textural features and the degree of microcracking of the material petrographic analysis. The petrographic analysis allowed to classify the rock as Ultramylonite feldspathic quartz. Macroscopically we can describe it as a rock of predominantly green. Their degree of microcracking settles around $1.8 / \text{mm}^2$.</p>	<p>The results of technological tests showed that the rock has good quality for ornamental use and coating even in places with large temperature ranges. The radon exhalation rate was within the limits of the United States Environmental Protection Agency. Within this context, the Ultramylonite feldspathic quartz proved to be very tough for use as coating materials.</p>

<p>10) AMARAL, P.G.Q. Evaluation of alterability and exhalation of radon gas in ornamental rocks. Ph. D. in Architecture and Urbanism, Educational Institution: University of São Paulo (USP/São Carlos), 2016. Keywords: Built environment; Dimension stone; Indoor contaminants; Radioactivity; Radon gas. Abstract – This thesis is based on the study of dimension stone in relation to aspects of changeability which will directly influence the increased addition of radon radioactive gas within an environment over time of use, a fact due to interactions with the environment where it is installed rock, as an example of its maintenance and cleaning, which will result in its aesthetic standard and quality of indoor air environment. Topics such as the quality of the related indoor air to the user's quality of life, natural radioactivity, the danger of radioactive radon gas into the atmosphere and dimension stone as composition and use in architecture are presented to foundation this Thesis and aiding in the understanding of tests.</p>	<p>For this, five ornamental stones were chosen and performed chemical tests changeability, technological characterization of assays and radiometric characterization tests to predict the behavior of a given rock in your environment to be applied in architecture. The attack of the HCl acid in the rocks decreased its strength and increased the amount of exhaled radon related directly to the direct increase in porosity, also increasing exhalation efficiency of the rock. The attack K(OH) alkali spite of reducing the resistance of the rock, and interfere with the porosity thereby decreasing the amount of radon vented in some cases.</p>	<p>This shows that even knowing the material and its contribution to the addition of Rn into the environment, with the passage of time the rock will increase your exhalation when not maintained can increase the level of accumulated radon in the environment. Thus, the thesis highlights the importance of knowing the material that will form the inside of the environment in order to prevent further damage to the health of the building and users, thus facilitating the understanding of these materials and how it interferes in the quality of the built environment.</p>
<p>11) CLARO, F. Protocol proposal for measuring radon concentrations from granitic rocks in marble works. Ph. D. in Electrical Engineering and Industrial Informatics, Federal Technological University of Paraná (UFTPR), 2016. Keywords: Natural Radioactivity; Granite; Radon; Marble Factory, Building Materials. Abstract – Naturally occurring radionuclides such as radon (^{222}Rn), its decay products and other elements from the radioactive series of uranium (^{238}U and ^{235}U) and thorium (^{232}Th) are an important source of human exposure to natural radioactivity. Worldwide the evaluation of radiobiological effects and risks to health from exposure of the population to natural radionuclides is a growing concern. Elements such as radon (^{222}Rn), the thoron (^{220}Rn), radio (^{226}Ra), thorium (^{232}Th) and potassium (^{40}K) can be found in materials commonly used in construction of houses and buildings. Thus, the radioactivity study from marbles and granites is important, given that under certain conditions these materials radioactivity levels can be hazardous requiring the implementation of mitigation measures for their use.</p>	<p>This study presents a technical protocol for the control of human exposure to natural radioactivity from granitic rocks in marble factories. The protocol was based on measurements of the ^{222}Rn and ^{220}Rn concentration in Brazilian granite rocks commonly nationally and exported. The ^{222}Rn and ^{220}Rn measurements were done using the AlphaGUARD (Saphymo GmbH) and RAD7 (Durrige Company) equipment's, respectively. For measures the samples of granite were sealed in glass jars for 40 days in order that the ^{226}Ra and ^{222}Rn radionuclides entered in secular equilibrium. The measurements were performed on Applied Nuclear Physics Laboratory at the Federal Technological University of Paraná. At the same time, solid-state nuclear track detectors CR-39 were installed in a marble factory environments located in Curitiba - Paraná for the evaluation of ^{222}Rn concentrations in workplaces. The CR-39 detectors were exposed for about 90 days and submitted to etching process.</p>	<p>The alpha particle tracks were observed using an optical microscope. The average ^{222}Rn concentrations of granite samples ranged from 32 Bq/m³ to 1,7 KBq/m³. The results obtained underscore the importance of this research in the data contribution to the development of national legislation that establishes limits of radioactivity values for marketing and use of granitic rocks. The results also contribute to the Brazilian granite meets the international standards that limit the acceptable radioactivity value for the import, export and transit the products such as marble and granite.</p>
<p>12) SILVA, F. D. O. Evaluation of color and exhalation rate of radon in granitic rocks through accelerated aging cycles. Ph. D. in Geology, Federal University of Ceará (UFC), 2016. Keywords: Radon; Accelerated Aging; Dimension Stones.</p>	<p>In the samples were conducted permeability, porosity, colorimetry, image analysis, petrographic and exhalation rate of radon, accompanied by aging tests on climate simulation chamber which simulates change situations of materials by weathering agents, accelerating wear and tear samples. The measurements were</p>	<p>The rocks have radon concentration values above the limits suggested by relevant international agencies (200-400 Bq/m³), with average values in the natural state, in 6149, 1619 and 866 Bq/m³ for Juparana Bordeaux, Branco Nevasca and Golden Arctic, respectively. The other aging cycles (50 and 100 cycles) showed an average increase of 0.8% for Juparana Bordeaux, 6.9% for White</p>

<p>Abstract – In this thesis are willing data used for the assessment of the analyzes performed on three types of dimension stone (Juparana Bordeaux, Branco Nevasca and Golden Ártico), in natural state and after several cycles of accelerated aging, correlating with the gas exhalation rate radon issued by the analyzed lithologies.</p>	<p>performed on samples in natural state, with 50 and 100 cycles of aging acceleration, where each cycle corresponds to variations in temperature and humidity in climatic simulation chamber, with the addition of an internal atmosphere of SO₂ with 25 concentration ppm. The results obtained during the tests were related to better analysis of the changes observed on the samples and the variation rate of exhalation radon emitted.</p>	<p>Blizzard and -23.87% for the Golden Arctic, with 50 cycles. From 50 to 100 cycles, there was reduction of 3.43% for Juparana Bordeaux and 22.15% for Branco Nevasca and an increase of 13.82% in the Golden Ártico. The porosity results in the natural state obtained values an average of 0.696% for Juparana Bordeaux, 0.919% for Branco Nevasca and 0.830% for Golden Ártico, and after 50 cycles of accelerated aging, obtained 0.621% to Juparana Bordeaux, 0.910% for Branco Nevasca and 0.840% for Golden Ártico. The permeability of the samples showed values in the natural state, in mDarcy, of 1,49E⁻⁴ to Juparana Bordeaux, 3,41E⁻⁴ to Branco Nevasca and 1,56E⁻⁴ for the Golden Ártico, and in 100 cycles, the results were 4,92E⁻⁴ to Juparana Bordeaux, 4,72E⁻⁴ to Branco Nevasca and 4,88E⁻⁴ for Golden Ártico. The image analysis and colorimetry showed that the samples visually showed minor variations in color during the accelerated aging cycles. With these results we it is considered that after accelerated aging cycles, the rocks studied showed an increase in most parameters assessed, especially in permeability, and a consequent increase in the rate of exhalation gas radon.</p>
<p>13) HAJJ, T.M. EL. Method for evaluating the use of ornamental rocks in interiors considering the exhalations of radon and thoron. Ph. D. in Mineral Engineering, University of São Paulo (USP), 2017. Keyword: Construction; Ornamental rocks; Radiation protection; Radon; Thoron. Abstract – Radioactive radon gas isotopes are generated from any soil or rock containing uranium and thorium in its composition. Thus, ornamental rocks used in construction are natural sources of radon and can generate indoor concentrations higher than those recommended internationally. As Brazil is an important producer and exporter of ornamental rocks; it was investigated in this research: an inexpensive and efficient way of sorting samples; correlations between gamma Index and the exhalations of radon and thoron; the influence of physical parameters on rock exhalations; Rn-222 diffusion length in rock plates; methods for reducing radon and thoron exhalations; preferential associations of uranium and thorium with other chemical elements and the proportions of radon and thoron in samples.</p>	<p>Forty (40) ornamental rocks samples collected in Brazil and Switzerland were studied and submitted to the following analysis: X-ray fluorescence, X-ray diffraction, scanning electron microscopy, thin section microscope analysis, pycnometry, moisture analysis, porosity, permeability, gamma spectrometry, neutron activation analysis, confocal microscopy, radon and thoron exhalation analysis using scintillation cell, radioactivity detection using a portable contamination detector and application of a waterproofing substance. Univariate and multivariate statistical analysis were conducted with Statistica 13 software.</p>	<p>The main results of the recommendations were as follows: use of a portable radiation detector as screening method for the selected samples; adopt gamma Index limit value of one (1) for ornamental rock plates; adopt total radon exhalation limit value of 150 Bq/hm²; use polishing and waterproof substance as surface treatment methods for reducing radon and thoron exhalations rates; minerals containing rare earth elements showed higher amount of associated U and Th; Th/U proportions in Brazilian samples (5 to 15) were higher than the global average (3 to 4).</p>
<p>14) LEAL NETO, A. Technological characterization of pegmatitic ornamental rocks from Seridó Oriental Paraibano – Pedra Lavrada/PB. Ph. D. in Geology, Federal University of Ceará (UFC), 2017. Keywords: Pegmatite; Technological Characterization; Resistance to Chemical Attack.</p>	<p>The results obtained in the technological characterization indicate that the investigated pegmatites can be used as ornamental rock, however, reservations regarding the use of White pegmatite are required, as a function of their performance in the technological tests related to physical indices, resistance to uniaxial compression and resistance to</p>	<p>The results of the colorimetric analysis showed that potassium hydroxide was the chemical reagent responsible for the highest chromatic variation observed in the White pegmatite ($\Delta E^*=7,78$) and for the pegmatites Capuccino ($\Delta E^*=8,55$) and Golden ($\Delta E^*=9,18$) color changes were caused by the action of citric acid. The values of radon exhalation in the studied rocks are considered high,</p>

<p>Abstract – The rocks studied are known commercially as Golden Fuji, Fuji White and Fuji Capuccino. They come from the same mining front located in the municipality of Pedra Lavrada/PB. They have a similar mineral composition consisting mainly of quartz, plagioclase, muscovite, microcline and garnet with contents between 5% and 34% and accessory minerals, with contents less than 5%, especially sericite, biotite and opaque. Petrographically correspond to pegmatites with isotropic structure, mineral granulation ranging from medium to very coarse, high degree of mineral microcracking, incipient to moderate mineral alteration and predominant concave-convex mineral contacts.</p>	<p>values lower than those suggested by current standards. The chemical solutions used affected the initial brightness intensity of the rocks in different proportions: potassium hydroxide caused a loss of 35,6% in the white pegmatite and citric acid induced a loss of brightness of 21,3% and 22,2%, in the Golden and Capuccino pegmatites, respectively.</p>	<p>mainly for the White pegmatite (215,0 Bq/m³), Capuccino (176,3 Bq/m³) and Golden (156,0 Bq/m³). The results for radon gas exhalation efficiency demonstrate that the amount of ²²²Rn effectively emanated in the rocks is small relative to the total amount of ²²²Rn generated. The simulation for indoor environments shows that the dosimetric values obtained for White pegmatite were 0,88 pCi/L; 0,73 pCi/L for the Capuccino pegmatite and 0,64 pCi/L for the Golden pegmatite revealing values below the dosimetric limit of 4,0 pCi/L. The results of the study of the thermal conductivity post-etching indicate that there was a reduction in the thermal conductivity values in the pegmatitic types studied (Capuccino: 4,1%, White: 3,4% and Golden: 2,2%) when compared to measurements of the thermal conductivities obtained before the action of the chemical reagents.</p>
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