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# Natural radionuclide distribution in Brazilian commercial granites

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**Abstract**

The dimension stones sector in Brazil produces several varieties of granites, marbles, slates and basalts. More than half of this production corresponds to around 200 different commercial types of granites with specific names, geographical and geological origins and mineral compositions. The well-known natural radioactivity present in rocks, where high radiation levels are associated with igneous rocks such as granite, can be used to determine their general petrologic features. This subject is important in environmental radiological protection, since granites are widely used as building and ornamental stones. In this paper, it is applied to correlate the petrographic characteristics of commercial granites with their corresponding dose rates for natural radioactivity. Amounts of thorium, uranium and potassium concentrations have been reported in several Brazilian commercial granite samples.


*Keywords:* Commercial granite; Potassium,Uranium and thorium concentrations; Petrologic signature; Annual effective dose rate

## 1. Introduction

Usually granites have a commercial name, petrographic and technological characterizations, and identification of the producer country (MMRO, 2002; ABIROCHAS, 2003; Pivko, 2003). Granites, usually suitable as building and ornamental materials for interior and exterior use, are hard natural stones that require harder tools to be cut, shaped and polished, compared to marble. Distinct types of commercial granites have different geological origins and mineralogical compositions and may be either magmatic or metamorphic rocks. Concerning their compositions, granites are mixtures of minerals of visible multicolored grains. One-color grains are typically encircled by grains of other colors, e.g. gray quartz is close to pink orthoclase, white plagioclase and dark mica. Every commercial granite contains feldspar (hardness 6 in Mohs Scale) of various colors: white, pink, red, yellow, brown, green and gray. Feldspar grains are typically not translucent and have a cleavage. Many granites, especially of light colors, contain quartz (hardness 7 in Mohs Scale) with gray (sometimes bluish) color and the grains are glassy translucent without cleavage. Further, there are dark minerals such as hornblende, pyroxene and biotite with black, dark green or dark brown colors. These minerals have larger specific gravity and lower hardness than feldspars and quartz.

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Some granite contains garnet showing near round shape and brown to dark-red color (Pivko, 2003).

Thorium, uranium and potassium concentrations of granitic rocks are intimately related to their mineral compositions and general petrologic features (Whitfield et al., 1959; Rogers and Ragland, 1961; Doventon and Prensky, 1992). Uranium and thorium in igneous and metamorphic rocks are usually found in a few accessory minerals such as apatite, sphene and zircon. Other highly radioactive minerals, like monazite, allanite, uraninite, thorite and pyrochlore, are widespread in nature, but they are very minor constituents of rocks. Uranium tends to be highly mobile near the surface whereas thorium is relatively stable. Uranium is easily oxidized to a water-soluble form and can be readily leached from pegmatites and granites and re-deposited in sediments at large distances from the source rock. Thorium is much less soluble than uranium and potassium and does not move except by mechanical means such as wind and erosion processes. Both thorium and uranium contents tend to be high in felsic rocks and to increase with alkalinity or acidity, with their highest concentrations found in pegmatites. The potassium content of rocks also increases with acidity. Potassium is usually found in potash feldspars, such as microcline and orthoclase, or in micas, like muscovite and biotite. Rocks that are free of these minerals have very low K-activity.

The petrologic features of granitic rocks associated with effects of weathering and metamorphism produce expressive alterations in the relationship between the natural radionuclides (Th, U, K, Th/U and Th/K). Consequently, the measurements of thorium, uranium and potassium concentrations of different granite samples result in individual differentiation sequences. From the $^{232}$Th, $^{238}$U and $^{40}$K activity concentrations obtained for each sample, it is possible to evaluate their respective dose rates in air, when these stones are used as tiling rocks (UNSCEAR, 1993, 2000; EC, 1999; Tzortzis et al., 2003; Anjos et al., 2004). These results are of great interest in the environmental radiological protection study, since granites are widely used as building and ornamental materials, including indoor covering.

Brazil is the sixth largest stone exporter in the world. Almost half of Brazilian production (47%) is concentrated in the small Espírito Santo State. Minas Gerais State is the second largest, and is responsible for the highest diversity of extracted minerals. The production from Minas Gerais and Bahia States correspond to 33% of the Brazilian production, whereas other ten states produce the other 20% (ABIROCHAS, 2003).

Using a NaI gamma-ray spectrometer to determine the concentration of natural radionuclides in an extensive selection of Brazilian samples, the annual effective dose rates and the gamma activity concentration index will be evaluated and compared to the limits proposed by United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR, 1993) and European Commission (EC, 1999), respectively. Additionally, the correlations between thorium, uranium and potassium will be shown in this paper with an aim to correlate the petrographic characteristics of commercial granites with their corresponding dose rates for natural radioactivity.

## 2. Material and methods

### *2.1. Sample collection and preparation*

Samples of 110 different types of the main Brazilian commercial granites were collected directly from the producers in ten Brazilian States: Espírito Santo (ES), Minas Gerais (MG), Bahia (BA), Rondônia (RO), Ceará (CE), Pernambuco (PE), Goiás (GO), Rio de Janeiro (RJ), São Paulo (SP) and Paraná (PR). We did so in order to avoid eventual misidentifications of the granites, due to the fact that in the stone markets there are examples in which different granites are sold as if they had the same commercial name or, on the other hand, the same granite has different names. Similarly it also occurs in the extraction zone that, sometimes, blocks of granites showing small heterogeneities, due to the distribution of multicolored grains, are extracted from same mining area (in some cases, from the same rock outcropping) are classified with different commercial names, although their basic mineralogical composition had not been changed. Thus, 95 of these granite samples collected were classified in accordance to their colors, sites of extraction and mineralogical compositions. In addition, a special attention was given to samples of brown granites, since it was recently reported in the literature (Tzortzis et al., 2003) that there is one type of Brazilian brown granite (*Café Brown*) which exceeds the acceptable dose criteria for the natural radioactivity of building materials recommended by the European Union (EC, 1999).

Fig. 1 show the localization of the Brazilian States where the granite samples were collected. For each type of the commercial granite, from three to five samples were measured. For brown granites, a set of ten different samples were measured.

In the Laboratory of Radioecology (LARA) of the Physics Institute of the Federal Fluminense University (IF-UFF), all rocks samples were first ground to powder. They were packed into cylindrical plastic containers, dry-weighed and sealed. About 150 g of sample material were used for the measurements of each granite sample. Before the measurements, the containers were kept sealed during 4 weeks, in order to reach the equilibrium of the $^{238}$U and $^{232}$Th series and their respective progeny. It was assumed that $^{220}$Rn and $^{222}$Rn could not escape from the sealed containers after closure.

### *2.2. Calibration and measurements by gamma ray spectrometry*

The measurements of the radioactivity concentrations were carried out at LARA, using the conventional