RADIOACTIVITY DOSAGE OF ORNAMENTAL GRANITIC ROCKS BASED ON CHEMICAL, MINERALOGICAL AND LITHOLOGICAL DATA

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ABSTRACT

One hundred samples of granitic rock were collected from granite traders in Belo Horizonte. Autoradiography, optical microscopy, diffractometry, and chemical analysis (X-ray spectrometry, X-ray fluorescence, neutron activation, gravimetry and electron probe microanalysis) were used to determine the mineral assemblages and lithotypes. Autoradiographic results for several samples showed the presence of monazite, allanite and zircon. Chemical analysis revealed concentrations of uranium of ≤30ppm, and thorium ≤130ppm. Higher concentrations generally correlated with high concentrations of light rare earths in silica-rich rocks of granitic composition. Calculations were made of radioactive doses for floor tiles in a standard room for samples with total concentration of uranium and thorium greater than 60ppm. On the basis of calculations of ^{232}Th, ^{40}K and ^{226}Ra from Th, K and U analysis, the doses calculated were between 0.11 and 0.34 mSv/year, which are much lower than the acceptable international exposure standard of 1.0 mSv/year.

Keywords: dosimetry, ornamental granite, autoradiography,

I INTRODUCTION

The radioactive study of Brazilian ornamental and finishing rocks carried out in this work is necessary as a parameter for a greater quality control of these rocks. The use of ornamental rocks in civil construction is an important economic activity, both in the domestic and foreign markets, with growing exportation that represents an important revenue source for Brazil (Ministério das Minas e Energia, 2001).

Most studies of ornamental and finishing rocks are concerned mainly with material resistance and their commercialization (Oliveira, 1998). Data about the radioactivity of these rocks is scarce (Salas *et al.*, 2000). Some of these rocks, mainly quartz-bearing plutonic rock (Salas *et al.*, 2002), present relatively high radioactive levels (Salas, 2003) resulting predominantly from uranium and thorium in monazite, allanite and zircon; and secondarily from uranium and thorium in titanite and apatite. Some of the radioactivity result from secondary uranium minerals and also from decay of the potassium radioisotope ^{40}K .

The development of this work is justified by the importance of the geographic distribution of these rocks in Brazil and their great economic potential. Several other products are derived from these rocks, including sand, clay, and metallic products (World Nuclear Association, 1998). Finally, since these rocks are considered U and Th rich, they have a great potential to future mining interests.

Materials and methods. The samples were obtained from granite traders at Belo Horizonte and included 100 different varieties of granites from the states of Minas Gerais and Espírito Santo in Brazil. Samples of processing waste from parent material weigh 2 kg each were requested. Samples were collected in a representative way from polished plates measuring 2 x 3 x 2m and weighing 300kg. This crushed material was sufficient to carry out all planned tests and analyses, as well as macroscopic and petrographical examinations, characterization of different lithotypes, and the determination of primary and secondary minerals (both radioactive and non-radioactive). In addition to preparation for examination, the samples were submitted for chemical determination of the main oxides using X-ray fluorescence, gravimetry, volumetry, atomic absorption and flame photometry. Rare earths in the samples were determined by energy of X-rays spectrometry. Thorium, radium and potassium analyses were performed by neutron parametric activation (INAA). The distribution of radioactive minerals in polished thin section was determined by autoradiography, and mineralogical studies were done using optical microscope and electron microprobe methods.

The results of the analyses and tests were sufficient to determine that a more detailed study involving geochemical characterization and radioactivity evaluations should be carried on a subset of 23 samples of those with uranium and thorium content over 60ppm. This sample group was also submitted to microanalysis.

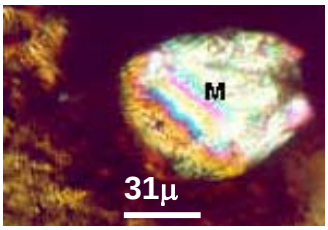
Petrographic and autoradiographic characterization. The samples are yellowish, gray and reddish, with sparse pink, whitish, lilac and brown spots. The macroscopic texture of these samples varies in grain size and homogeneity. Some are oriented gneiss and migmatite, commercially called “moved rocks”. Such samples contain regular or irregular layers attributed to deformation provoked by tectonic efforts, which occurred either during or after intrusion.

The primary mineralogical composition determined using a petrographic microscope revealed that more than 80% of each sample is composed of quartz (9.7 – 33.4%), k-feldspar (24.3 – 51.3%) and sodic plagioclase (24.8 – 41.4%). Another common mineral in these rocks is mica (predominantly biotite, muscovite to a lesser extent). Secondary minerals resulting from varying degrees of alteration of the primary minerals are represented mainly by clay minerals (kaolinit and montmorillonite), sericite, carbonates, iron oxides minerals and chlorite.

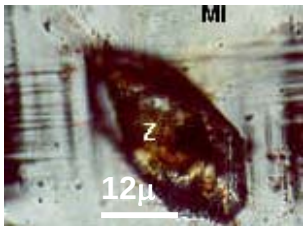
Accessory minerals, generally with contents lower than 1%, but occasionally higher, are mostly zircon, monazite, apatite, opaque minerals (magnetite, hematite/martite, ilmenite, pyrite, pyrrhotite, goethite and chalcopyrite), allanite, rutile, fluorite, epidote, xenotime, bastnaesite, and gummite (Figure 1).

A predominantly brown-colored syenite sample (CG-90) (Table 1) presented a different mineralogical composition, showing a predominance of k-feldspar (73.4%), along with sparse, quartz and plagioclase, and abundant augite, hornblende and biotite.

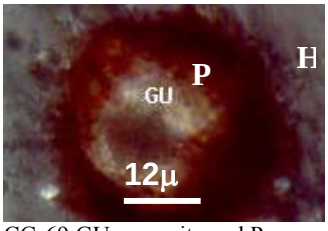
Petrographic microscopic characterization was carried out to quantitatively determine the mineralogical constituents of the 100 samples. For the lithographic classification, we used the QAP diagram of Streckeisen (1976) as presented by Le Maitre (1989). On the basis of mineralogy, the samples were classified mainly as monzogranite, with some other lithologic types such as syeno-granite, monzonite, quartz monzonite, granodiorite, monzodiorite monzogabbro, quartz gabbro and syenite.



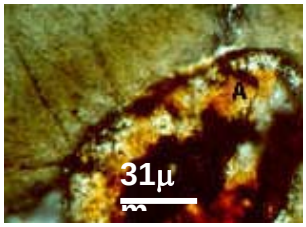
CG-23 M-monazite and B-biotite



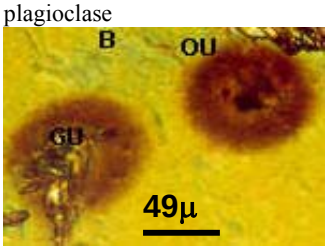
CG-31 Z-zircon and MI-microcline



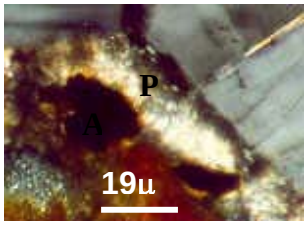
CG-69 GU-gummite and P-plagioclase



CG-90 A-allanite and H-hornblende



CG-90 G-gummite, OU- oxide of uranium and B-biotite



CG-93 A-allanite and P-plagioclase

Figure 1 - Photomicrography of radioactive minerals in granite, in section using polarized light

Table 1 – Ornamental granitic rocks combined concentration with U_3O_8 and ThO_2 above 60 ppm

Sample	Classification		Density Kg/m^{-3}	U_3O_8 (ppm)	ThO_2 (ppm)	^{232}Th	^{226}Ra	^{40}K	H_{ET} mSv/ year
	Streckeisen (1976) and quoted by Le Maitre (1989)	La Roche (1964)				Bq.kg^{-1}			
Ceramic			1000			10	15	50	0.0068
CG-02	Monzo-granite	Syenogranite	2515	4	94	380	10	1600	0.21
CG-03	Syeno-granite	Syenogranite /Alkali Granite	2698	5	100	410	170	1600	0.28
CG-20	Monzo-granite	Alkali Granite /Syenitic	2653	2	70	240	222	1700	0.20
CG-23	Monzo-granite	Alkali Granite	2602	6	56	230	80	1700	0.16
CG-24	Monzo-granite	Syenogranite	2711	5	57	230	10	1500	0.14
CG-30	Monzo-granite	Alkali Granite	2645	20	59	240	400	1600	0.24
CG-31	Monzo-granite	Alkali Granite / Syenogranite	2618	5	76	310	30	1600	0.19
CG-33	Monzo-granite	Alkali Granite / Syenogranite	2399	6	74	300	222	1700	0.22
CG-35	Monzo-granite	Alkali Granite	2600	16	55	220	440	1300	0.24
CG-37	Syeno-granite	Alkali Granite / Syenogranite	2533	1	66	270	10	1800	0.16
CG-42	Gneiss Monzo-granite	Gneiss Syenogranite	2936	4	130	530	140	1600	0.34
CG-46	Monzo-granite	Alkali Granite	2595	5	58	240	210	1600	0.19
CG-52	Gneiss Monzo-granite	Gneiss Alkali Granite	2670	8	67	270	60	1300	0.17
CG-63	Quartz Monzonite	Alkali Granite	2433	5	74	300	20	1500	0.17
CG-66	Monzo-granite	Alkali Granite / Syenogranite	2536	5	66	270	150	1500	0.19
CG-69	Gneiss Monzo-granite	Gneiss Alkali Granite / Syenogranite	2587	30	44	180	10	1300	0.11
CG-74	Gneiss Quartz Monzonite	Gneiss Quartz Syenite	2762	4	94	380	90	1700	0.24
CG-78	Gneiss Quartz Monzonite	Gneiss Alkali Granite	2527	7	66	270	220	1000	0.20
CG-90	Syenite	Syenite	2521	5	76	310	600	2300	0.33
CG-93	Syeno-granite	Syenogranite	2823	3	81	330	10	1700	0.20
CG-97	Monzo-granite	Monzogranite / Syenogranite	2660	3	65	270	20	1600	0.16
CG-98	Gneiss Monzo-granite	Gneiss Syenogranite	2691	4	105	430	10	1800	0.25
CG-100	Monzo-granite	Syenogranite	2715	4	58	240	10	1500	0.15

Note: The majority of the samples are peraluminous, excepted CG-63, which is peralkaline and CG-90, which is metaluminous.
Peraluminous ⇒ AL > Na and K oxides, Peralkaline ⇒ Al < Na and K oxides, Metaluminous ⇒ Al > Na and K oxides but < Na, K, Ca oxides combined and Subaluminous ⇒ few to ϕ Al oxide

Autoradiographic tests using photographic plates allowed the detection of radiation with energy on the order of 6 MeV in contents higher than 100ppm. Localization of radioactive minerals, were shown by the shapes, sizes and intensities of irregularly distributed darkened spots. These spots were analyzed using optical microscopy and electron microprobe analysis and were found to mostly contain grains of monazite, allanite and zircon in the majority of the samples. These minerals contain radioactive elements which produced discolored haloes or radial fractures in the minerals they are included in.

Geochemical characterization. Chemical analysis of the main oxides in the radioactive subset by X-ray fluorescence, atomic absorption, volumetry and flame photometry determined the contents as SiO_2 (59.7 – 76.5%), Al_2O_3 (11.7 – 15.6%), K_2O (3.8 – 7.4%), Na_2O (1.91 – 4.52%) and Fe_2O_3 (1 - 8%). Other oxides analyzed such as CaO, MgO and TiO_2, presented amounts less than 1.33%.

On the basis of neutron activation analysis of the main sample group, uranium varied between 1 to 30ppm and thorium between 44 to 130ppm. The presence of these oxides is attributed mainly to the presence of monazite, allanite and zircon.

Uranium and thorium contents in these samples were determined by X-ray energy spectrometry (EDS), and in general they correlate with light rare earths: La_2O_3 (50 - 300ppm), Ce_2O_3 (30 - 580ppm), Pr_6O_{11} (20 - 580ppm) and Nd_2O_3 (30 - 200 ppm).

X-ray fluorescence analysis of all sample results were plotted on a La Roche (1964) diagram to provide a lithogeochemical classification. Using this method, samples were classified as syenogranite and alkali granite, along with monzogranite, quartz syenite and nepheline syenite (Table 1). Oxide results, Al_2O_3 (A), CaO (C), K_2O (K) and Na_2O (N), were also used to determine aluminum saturation parameters in these rocks as defined by diagrams of Shand (1927). It was observed that in this sample group there occurred mainly peraluminous rocks, with the exception of sample CG-63 with peralkaline composition, and sample CG-90 with metaluminous composition (Table 1).

II RADIOACTIVITY EVALUATION

To evaluate the radioactivity in the ornamental granitic rocks with uranium and thorium concentrations greater than 60 ppm, our model of a "standard room" measured 4 x 4 x 3 m, with four brick walls (two 10cm and two 20cm thick, simulating both internal and external house walls), flat roof (12 cm thick) and ornamental granitic rock flooring (2 cm thick). We also considered a ventilation rate of 0.7 h^{-1}, room residence time of 7,008 hours per year, equivalent to 80% occupation during the year, and brick size 20 x 10 x 5 cm.

In calculations of the dose rate (H_{ET}) for granitic samples in relation to a reference ceramic (Ginjaar, 1985), the density in $kg.m^{-3}$ and the dose rates of ^{232}Th, ^{226}Ra and ^{40}K, given in Table 1, obtained through neutron activation analysis and converted in $Bq.kg^{-1}$.

The model for the "standard room" of Castro & Senne (1995) was used for the calculation dose of both ceramic and granite samples. However, the rates of equivalent doses were determined by difference of the doses due to their gamma radiation. The yearly effective dose, H_{E_γ}, was calculated using the following expression:

$$H_{E_\gamma} = T.b.\overset{o}{D} \qquad (1)$$

Where:

T = occupation time in the "standard room" in a year: T = 7008 $h.^{a-1}$;

b = conversion factor of the absorbed dose in air to the effective dose equivalent for terrestrial radiation: b = 0.7 $Sv.Gy^{-1}$

$\overset{o}{D}$ = air absorption dose rate [Gy.h^{-1}].

Air absorption dose rate, $\overset{o}{D}$, is the sum of the contributions of the different radionuclides present in the construction material:

$$\overset{o}{D} = \sum_i q_x . C_{x,i} . m_i \quad (2)$$

Where:

q_x = concentration conversion factor of radionuclide *x* present in building material with air dose absorption rate 1 meter away from the surface of the material [Gy.h^{-1}.Bq^{-1}.kg],

$C_{x,i}$ = radionuclide *x* concentration in material *i* [Bq.kg^{-1}] and

m_i = material *i* relative mass in "standard room"

Yearly equivalent dose, $H_{E_{Rn}}$, due to radon, a decay product present in construction material, was calculated through the following expression:

$$H_{E_{Rn}} = T \cdot r_{Rn} \cdot F_{Rn} \cdot C_{Rn,i} \quad (3)$$

Where :

T = permanence time in the "standard room" in a year: T = 7008 h.$^{a-1}$;

r_{Rn} = conversion factor for the equivalent balance of the concentration of the decay products of radon in the "standard room" with equivalent dose rate [mSv.m^{-3}.h^{-1}.Bq^{-1}];

F_{Rn} = is the equilibrium factor of the concentration of radon decay products in the outside air;

$C_{Rn,i}$ = radon concentration resulting from ^{226}Ra in construction material *i* in [Bq.m^{-3}] given by:

$$C_{Rn,i} = S_i \cdot \frac{A}{V} \cdot \frac{1}{\lambda_v + \lambda_{Rn}} \cdot f_{Rn} \cdot C_{Ra,i} \quad (4)$$

Where:

S_i = relative surface of material *i* in our "standard room";

A = area of material *i* in "standard room" [m^2];

V = volume of "standard room" [m^3];

λ_v = ventilation rate of "standard room" [h^{-1}];

λ_{Rn} = decay rate of Rn [h^{-1}];

$f_{Rn,i}$ = emission rate of radon per m^2 of material *i*, and per unity of ^{226}Ra concentration [kg.m^{-2}.h^{-1}] and

$C_{Ra,i}$ = concentration of ^{226}Ra in material *i* [Bq.kg^{-1}]

The emission rate of radon, $f_{Rn,i}$, is calculated through the following expression:

$$f_{Rn,i} = \lambda_{Rn} . \rho_i . \eta_i . l_i . \mathrm{tgh}\,(L_i / l_i) \quad (5)$$

Where :

ρ_i = density of construction material *i* [kg.m^{-3}];

η_i = emission factor of construction material *i* (indicates the fraction of radon that reaches the material pores);

l_i = diffusion effective length in material *i* [m] and

L_i = half thickness of material *i* used [m]

and where "tgh" means hyperbolic tangent function

The l_i factor is given by:

$$l_i = \sqrt{\frac{K_i}{\lambda_{Rn}.\varepsilon_i}} \qquad (6)$$

Where :

K_i = diffusion coefficient of radon in material *i* [$m^2.h^{-1}$] and

ε_i = material *i* porosity.

The calculation of thoron (Tn), a thorium decay product, is analogous to the calculation of radon, substituting Tn for Rn and Th for Ra.

Finally, the calculation of dose increase due to the use of granite instead of ceramic flooring is given by:

$$H_{ET} = (H_{E\gamma} + H_{E_{Rn}} + H_{E_{Tn}}) - H_{ET}\,ceramic \qquad (7)$$

The rate of dose and increase in mSv/year, due to the use of granitic flooring in the "standard room". In these calculations, the following values of parameters and constants were used:

B = 0.7 Sv Gy^{-12}

$Q_k = 54 \times 10^{-12}$ $Gy.h^{-1}.Bq^{-1}.kg$

$Q_{Th} = 890 \times 10^{-12}$ $Gy.\ h^{-1}.Bq^{-1}.kg$

$Q_{Ra} = 620 \times 10^{-12}$ $Gy.h^{-1}.Bq^{-1}.kg$

$r_{Rn} = 8.7x10^{-6}$ $mSv.m^{-3}.h^{-1}.Bq^{-1}$

$F_{Rn} = 0.4$ for a ventilation rate of 0.7 h^{-1}

$\lambda_{Rn} = 7.56 \times 10^{-3} h^{-1}$

$S_G = 0.20$; A = 16 m^2; V = 48 m^3; $\lambda_V = 0.7\ h^{-1}$, $M_G = 0.20$;

$l_G = 0.01$; $\frac{K_G}{\in_i} = 1.8x10^{-4} m^2 h^{-1}$

Where: subscript "G" refers to granitic material.

The dose rates of these samples (Table 1 and Figure 2) were in the range 0.11 – 0.34 mSv/year, with most samples with rates lower than 0.20 mSv/year. The largest dose rates occur in monzo-granite and syeno-granite, as defined by Streckeisen (1976), Le Maitre (1989), which correspond to alkali granite, alkali granite / syenogranite and syenogranite in the La Roche (1964) classification.

The reference dose rate was 0.0068 mSv/year as obtained from the data of Ginjaar (1985). An increased dose rate due to the use of granitic rocks instead of ceramic flooring was lower than 0.01 mSv/year for all samples, however.

Using the radionuclides ^{232}Th, ^{226}Ra and ^{40}K from sample CG-90 of the radioactive subset sample group, it was hypothetically determined 3 conditions in the "standard room" based on the spreadsheet: a) total use of granite (m = 100) as building material in the whole "standard room" in the same conditions of the works of Sharaf *et al.* (1999), Kumar *et al.* (1999), Amrani & Tahtat (2001), Rizzo *et al.* (2001) and Kovler *et al.*(2002) resulted in H (gamma) = 3.8 mSv/year and total dose rate of 9.04 mSv/year; b) no ventilation ($\lambda_v = 0$) and granite flooring of 5% (m=5) of the total construction material resulted in H (gamma) = 0.189 mSv/year and total dose rate of 5.44 mSv/year; and c) ventilation ($\lambda_v = 0.7$) resulted in yearly dose rate of 0.33 mSv/year. This last condition was used in the calculation of the total doses of all samples with uranium and thorium contents over 60ppm, which yielded doses of 0.11 – 0.34 mSv/year.

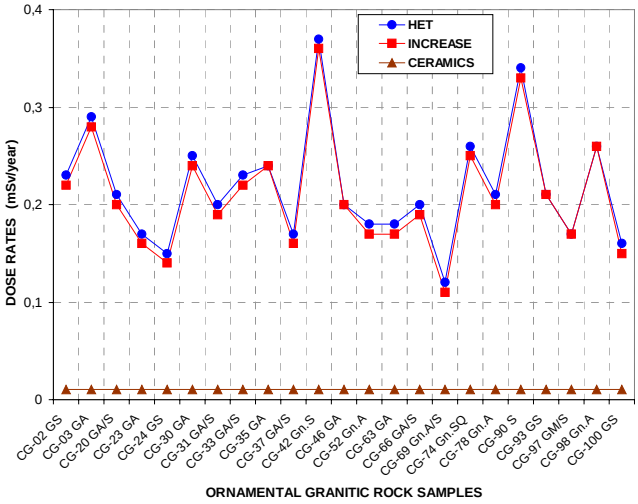


Figure 2 – Increase in dose rates for granitic rock (GA- alkali granite, Gn.A-gneiss alkali granite, GA/S- alkali granite / syenogranite, Gn.A/S-gneiss alkali granite / syenogranite, GS- syenogranite, Gn.S-gneiss syenogranite, GM- monzonite granite, S-syenite and Gn.SQ- gneiss quartz syenite), according to La Roche (1964) instead of ceramics in samples with total radioactive elements (U_3O_8 and ThO_2) over 60ppm.

III CONCLUSIONS

The 100 samples classified in the QAP (quartz, k-feldspar and plagioclase) diagram of Streckeisen (1976) quoted by Le Maitre (1989) are predominantly monzo-granite (73%) with lesser groups of granodiorite (9%), quartz monzonite (6%), monzonite (4%), syeno-granite (3 %), syenite (3 %), monzodiorite/monzogabbro (1%) and quartz gabbro (1%).

Most samples were classified according to La Roche (1964) as alkali granite, were alkali granite/syenogranite and syenogranite in comparison to those classified by Streckeisen diagram (1976) Le Maitre (1989) as monzo-granite and rarely syeno-granite. The remaining samples were characterized by geochemistry according to La Roche (1964) as being syenite and monzo-granite, very similar to syenogranite and monzonites and monzonite and granodiorite in the mineralogical-petrographical characterization of Streckeisen (1976).

Results from the calculation of increase dose from the granitic rock is still below harmful level to the public as the doses are less than the international standard of exposure to radioactivity, at 1.0 mSv/year, ("National Council on Radiation Protection and Measurements", 1987).

It is observed that the dose rate produced by radon and thoron derived from the use of granitic rock is considerably increased when the relative surface of material *i* in the "standard room" [m^2] presents larger percentages and also when there is little or no ventilation [h^{-1}].

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