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Evaluation of the radiation shielding properties of ornamental rocks produced in Brazil: A Monte Carlo approach

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ABSTRACT

In this work, the Monte Carlo MCNPX (2.7.0) code was used to evaluate the radioprotective properties of ten ornamental granitic samples produced in Brazil. For each sample of granite, the mass attenuation coefficient (μ/ρ) and half-value layer (HVL) were evaluated using the photon energy emitted by the following radioisotopes: ²⁴¹Am (59,5 keV), ¹³³Ba (356 keV), ¹³⁷Cs (662 keV), ⁶⁰Co (1250 keV), and ²²Na (1274 keV). The MCNPX results showed agreement with the values obtained by using the XCOM database, as well as with theoretical and experimental results, available on the literature. The computational model built in this work can be used by the scientific community interested in parameters involving new materials for gamma radiation shielding, which has been used in different areas of the nuclear sciences.

Keywords: Granitic rocks, Monte Carlo simulation, gamma radiation.



1. INTRODUCTION

Nowadays, all around the world, the use of ionizing radiation with different energies is becoming widely applied in different industry sectors, associated with the nuclear energy production and medicine [1]. Together with the increment in the use of ionizing radiation, the need to investigate new shielding methods also increases, intending to guarantee the radiological protection and personal exposures control. Therefore, it is fundamental to study the properties of new shielding materials against ionizing radiation.

Different types of materials (barites, sands, concretes, etc.) are used as shielding in areas where the ionizing radiation is applied. In general, they are associated with other construction materials such as bricks, plaster, and cement, for example [2].

The specialized literature presents Monte Carlo simulation studies in which the mass attenuation coefficient is evaluated for the gamma irradiated concrete containing barite [3, 4, 5]. In other studies, shields made with cement and mortar were analyzed [6, 7], as well as different types of rocks [8].

In addition to the evaluated composites, granitic materials are investigated to act as radiation shielding due to their high tenacity, lucrativeness, abundance, and mineral cohesion [9]. Brazil produces and exports, annually, thousands of tons of ornamental granitic rocks, which are applied in different fields, mainly in the construction [10]. These rocks are mineral aggregation formed through the magma cooling. There are two processes which may occur: 1. Plutonic rocks with phaneritic texture - cooling of these rocks at a depth of a few kilometers, and 2. Volcanic rocks with aphanitic texture - when magma cooling occurs on the surface (or close to it) [11].

Granite is a well-known type of igneous rock, used as a construction material, and its properties have been analyzing to be used as radiation shielding material [2, 12]. A vast number of both experimental and theoretical studies about the shielding properties of granite have been published. Akkurt *et al.* (2010) [3] studied the shielding properties of Indian granite. Another study, developed by Najam *et al.* (2016) [13], a Monte Carlo simulation was employed to determine a set of linear attenuation coefficient, as well as mass attenuation coefficient, for various types of granite used in the construction, using a gamma irradiation source.

In the present study, the attenuation properties of ten types of granitic rocks, from three different Brazilian states, were evaluated: 4 samples from the state of Ceará, 3 samples from Espírito Santo, and 3 samples from Mato Grosso. The results were obtained using the Monte Carlo MCNPX code [14] and the XCOM database [15]. The granitic rocks were exposed to different gamma radiation energies, emitted by radioisotopes such as ^{241}Am (59.5 keV), ^{133}Ba (356 keV), ^{137}Cs (662 keV), ^{22}Na (1274 keV), and ^{60}Co (1250 keV), which are widely used in different scientific and industrial fields. The obtained results were compared to experimental data from the literature and XCOM database, in order to evaluate a set of ornamental granitic rocks to use as shielding materials.

2. MATERIALS AND METHODS

In this study, the radiological protection properties of ten types of granitic samples, from three different Brazilian states (Ceará, Espírito Santo, and Mato Grosso), were evaluated. The MCNPX 2.7.0 Monte Carlo radiation transport code [14] was used to the calculations. The composition ($w\%$) and density of the ornamental rocks, utilized in this work (Silva *et al.* 2009 [16], Mattos *et al.* 2013 [17], Saar *et al.* 2015 [18], Pereira *et al.* 2019 [10], Santos *et al.* 2022 [19]), are presented in Table 1. Then, the obtained results for the set of granitic rocks were compared with different types of concrete used as shielding materials.

Table 1: Composition (w%) and density of the ornamental rocks utilized in this work [10, 16-19]

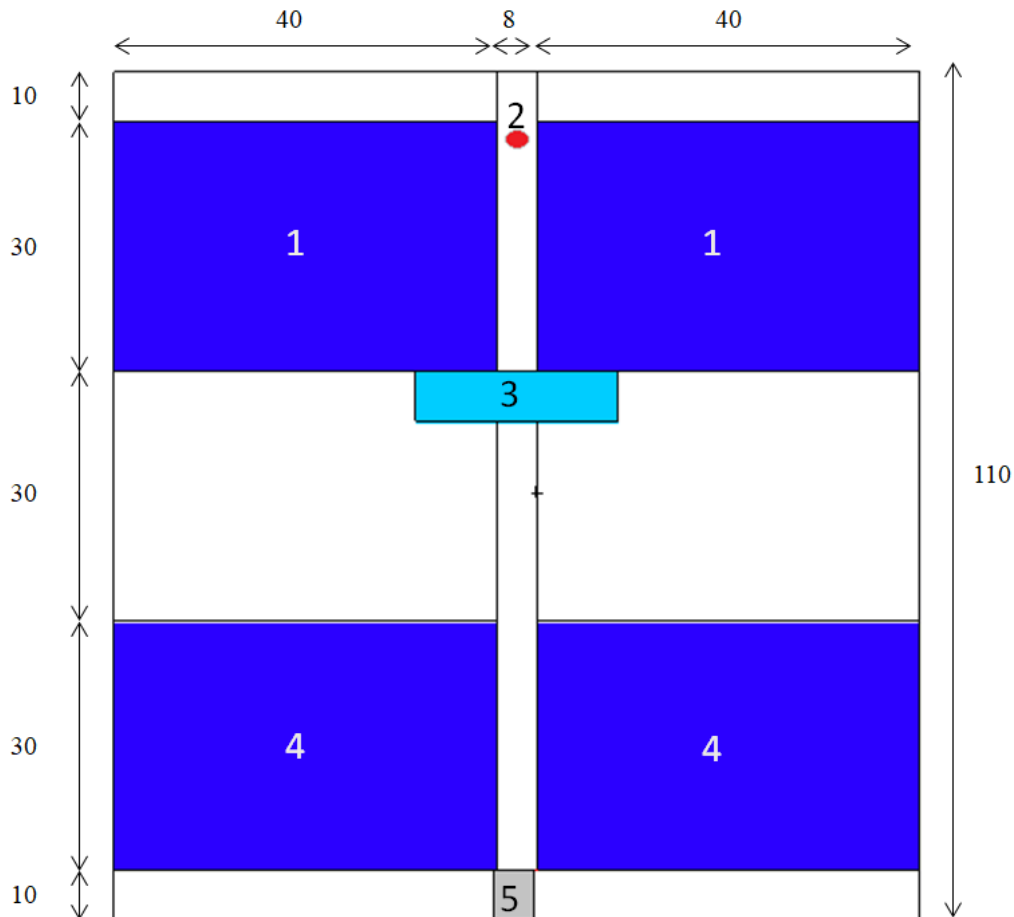
Common names	Brazilian state	Si	Ti	Al	Fe	Mn	Mg	Ca	Na	K	P	O	Density (g/cm³)
<i>Rosa Iracema</i>		35	0.5	3.59	0.63	0.01	0.21	0.60	1.15	2.28	0.02	56.41	2.62
<i>Rosa Olinda</i>	Ceará	33.88	0.14	3.68	1.27	0.03	0.23	1.00	1.22	2.03	0.01	56.48	2.63
<i>Branco Savana</i>		35.85	0.03	3.20	0.62	0.01	0.18	0.41	1.15	1.86	0.002	56.65	2.61
<i>Branco Cristal Quartzo</i>		33.56	0.14	3.75	0.75	0.01	0.12	0.95	1.35	2.49	0.01	56.84	2.62
<i>Giallo São Francisco</i>		34.17	0.14	3.31	0.83	0.02	0.19	0.61	1.35	2.28	0.03	56.74	2.64
<i>Branco Dallas</i>	Espírito Santo	33.34	0.15	4.02	0.87	0.02	0.19	0.59	1.05	2.61	0.03	57.09	2.63
<i>Branco Marfim</i>		34.20	0.13	3.75	0.74	0.04	0.21	0.62	1.14	2.28	0.02	56.85	2.63
<i>Granito Sararé</i>		22.34	1.61	4.94	0.37	0.08	0.21	4.65	1.25	1.33	0.12	57.70	2.59
<i>Anfibolito Canaã</i>	Mato Grosso	22.35	0.81	3.98	4.64	0.10	4.72	8.16	0.76	0.19	0.03	45.76	3.12
<i>Diabásio Salto do Céu</i>		34.98	0.04	3.74	0.29	0.01	0.03	0.45	1.71	1.85	0.004	56.87	2.85

2.1. Computational modeling of the exposure scenarios using MCNPX Monte Carlo code

The MCNPX 2.7.0 radiation transport code [14] is widely used by the scientific community to model the radiation interaction with materials, and to obtain information about radiological protection. Because of this, the code was selected to evaluate the shielding properties of the different types of granite.

In this study, a punctual radiation source emitting photons isotropically was considered. The monoenergetic photons emitted in the source representing the predominant energy of common radioisotopes had the following values: ^{241}Am (59.5 keV), ^{133}Ba (356 keV), ^{137}Cs (662 keV), ^{60}Co (1250 keV), and ^{22}Na (1274 keV). Ten types of granitic rocks and concrete were used in the analysis, being among the most used as shields. Figure 1 show the scheme used to obtain the linear attenuation coefficients. The following parts are presented in the scheme: four lead blocks used to collimate the beam, named as primary collimators (1), and secondary collimators (4), made of lead (density = 11.35 g / cm³, composition: Pb (100%)), gamma radiation source (2), and the granitic rock sample (3) (density, and composition Table 1), acting as an attenuator material, localized between the source and the detector (5) (air density = 0.001205 g / cm³, composition: C (0.0124%), N (75.53%), O (23.18%), and Ar (1.28%)). The detector was defined as an atmospheric cell of air localized in the exit of the second collimation pair.

Figure 1: MCNP view of the geometrical setting. Dimensions (in cm) of the geometry used to represent the scheme used to obtain the linear attenuation coefficients



Source: Author

The results were obtained by using the modified Tally *F4 (MeV/cm²/source-particle), aiming to register the energy flux of the photons in the detector area. The ENDF/B-VIII cross-section library was used, which considers the photoelectric absorption, Compton scattering and pair formation. Statistical uncertainties were reduced, in each simulation, by including 1E8 histories of particles, which provided relative uncertainty values less than 0.1%.

2.2. Mass attenuation coefficient

For a beam of monoenergetic photons, the intensity (I_x) decreases as the beam propagates through material, which can be described by the Lambert-Beer law represented in Equation 1 [2],

$$I_x = I_0 \cdot e^{-\mu x} \quad (1)$$

where x represents the thickness of the material; μ is the linear attenuation coefficient, which represents the cross-section of interaction between each photon and the material, depending on the density and the chemical composition of the sample; and I_0 represents the initial intensity of the radiation. The mass attenuation coefficient (μ/ρ) for photons in an attenuator medium is given by Equation 2 [20],

$$\mu/\rho = \ln(I_0 / I_x) / \rho x \quad (2)$$

where ρ is the density of the attenuator material and x is its thickness.

By using the modified Tally *F4 (MeV/cm²/source-particle) of the MCNPX code, and applying the Equation 2, it was possible to obtain μ/ρ . The half-value layer (HVL) was calculated from Equation 2 ($HVL = \ln 2 / \mu$) [21].

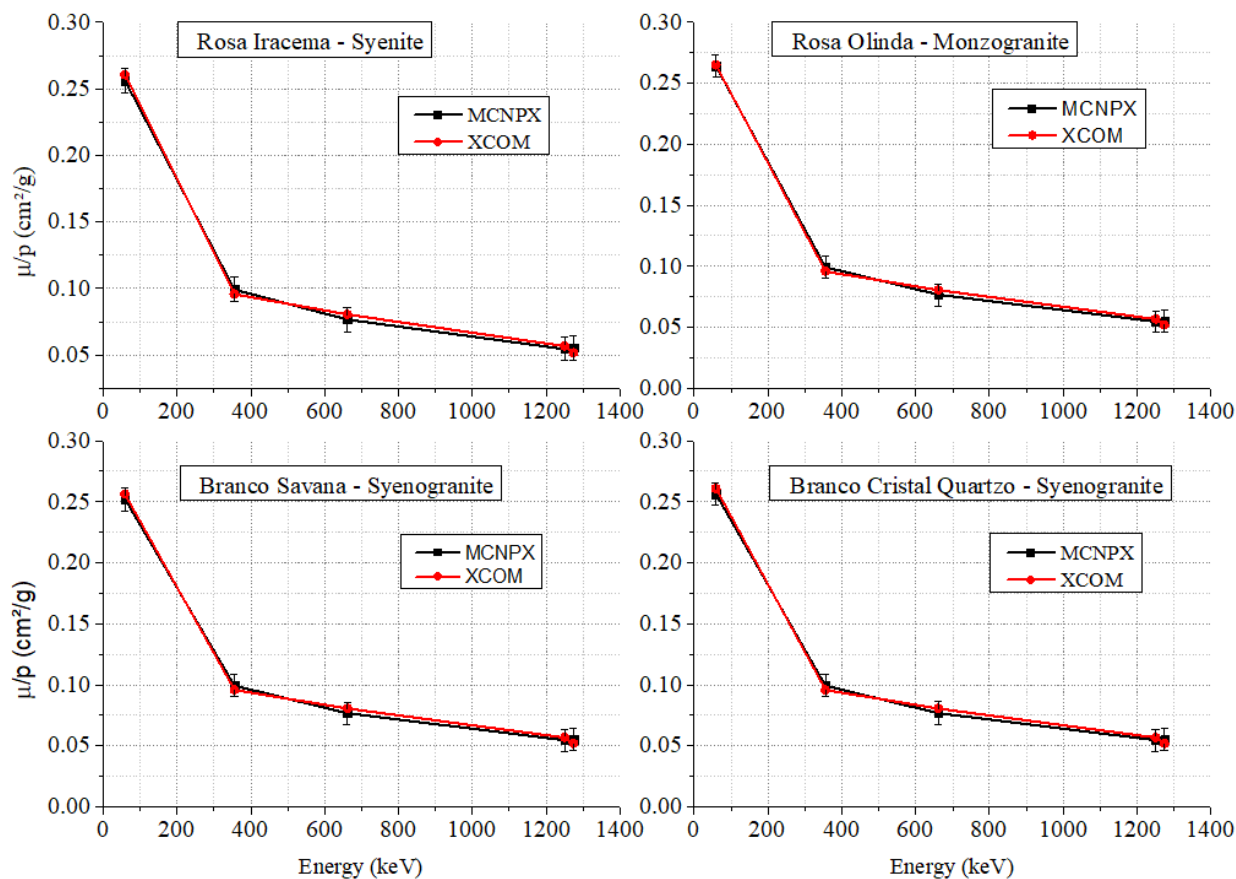
In this work, the main radiological attenuation properties such as μ/ρ and HVL were obtained for a range of energy from 59.5 keV to 1270 keV using the MCNPX code and XCOM database. The main chemical and physical properties of the studied materials were taken from a recent publication of our research group [19].

The XCOM database provides a broad photon cross section database in which the parameters specification and the input to the calculations are flexible and easy to change. Using this tool, each sample of granite was defined by its elemental composition fractions. The results obtained from XCOM, together with experimental data available on the literature, were compared with the MCNPX simulations.

3. RESULTS AND DISCUSSION

The main results for the mass attenuation coefficients are presented in Figures 2, 3 and 4.

Figure 2: Comparison between the mass attenuation coefficients for the rocks from Ceará State. They were obtained using MCNPX and the values from the XCOM database



Increasing the photon energy implicated in a reduction of the μ/ρ values, being the highest value obtained for the photon energy equal to 59.5 keV. These variations are directly connected to the photon interaction processes. Figures 2-4 show a strong dependence of the μ/ρ for the lower range of energy and, in the statistical uncertainty limits, the results from the MCNPX simulations agree with the results obtained by using the XCOM database for all the granitic rock samples.

Figure 3: Comparison between the mass attenuation coefficients for the rocks from Espírito Santo State. They were obtained using MCNPX and the values from the XCOM database

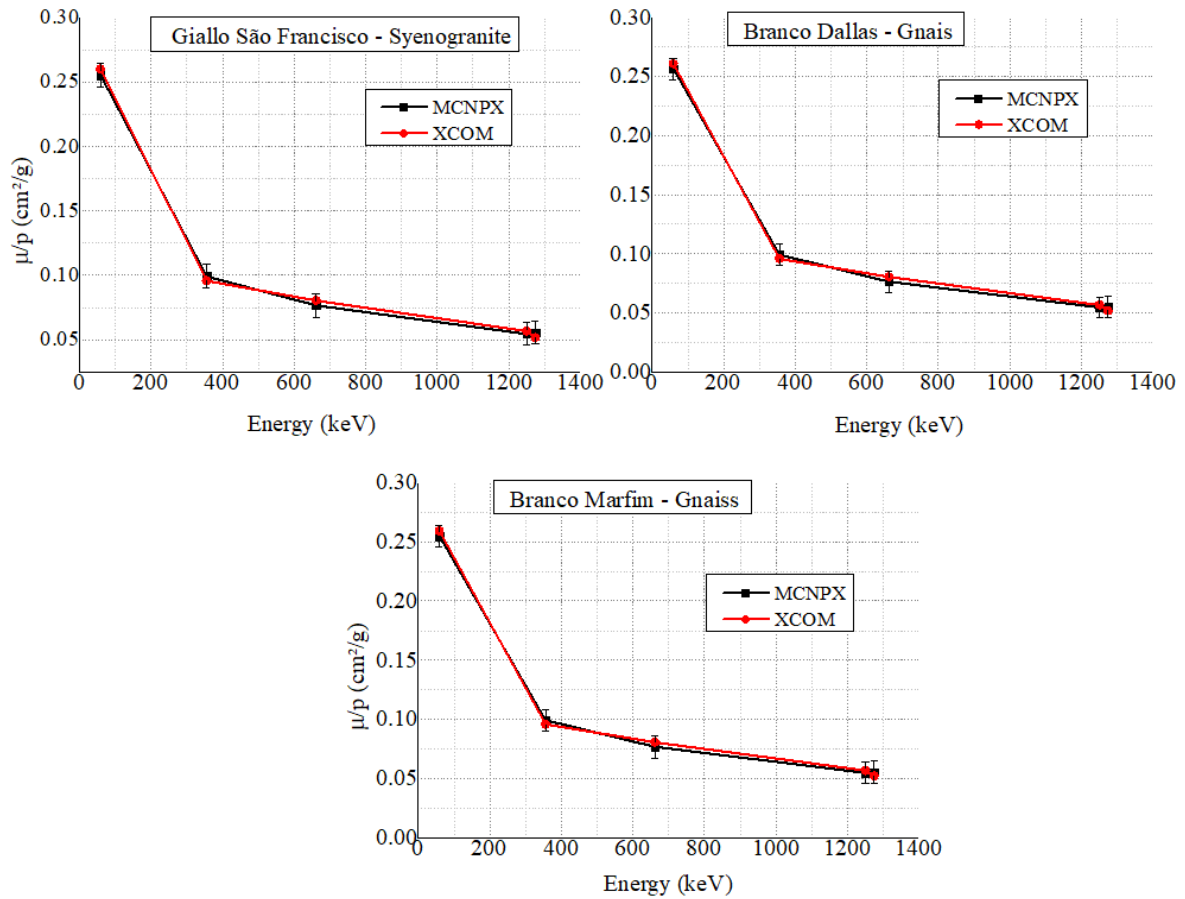


Figure 4: Comparison between the mass attenuation coefficients for the rocks from Mato Grosso State. They were obtained using MCNPX and the values from the XCOM database

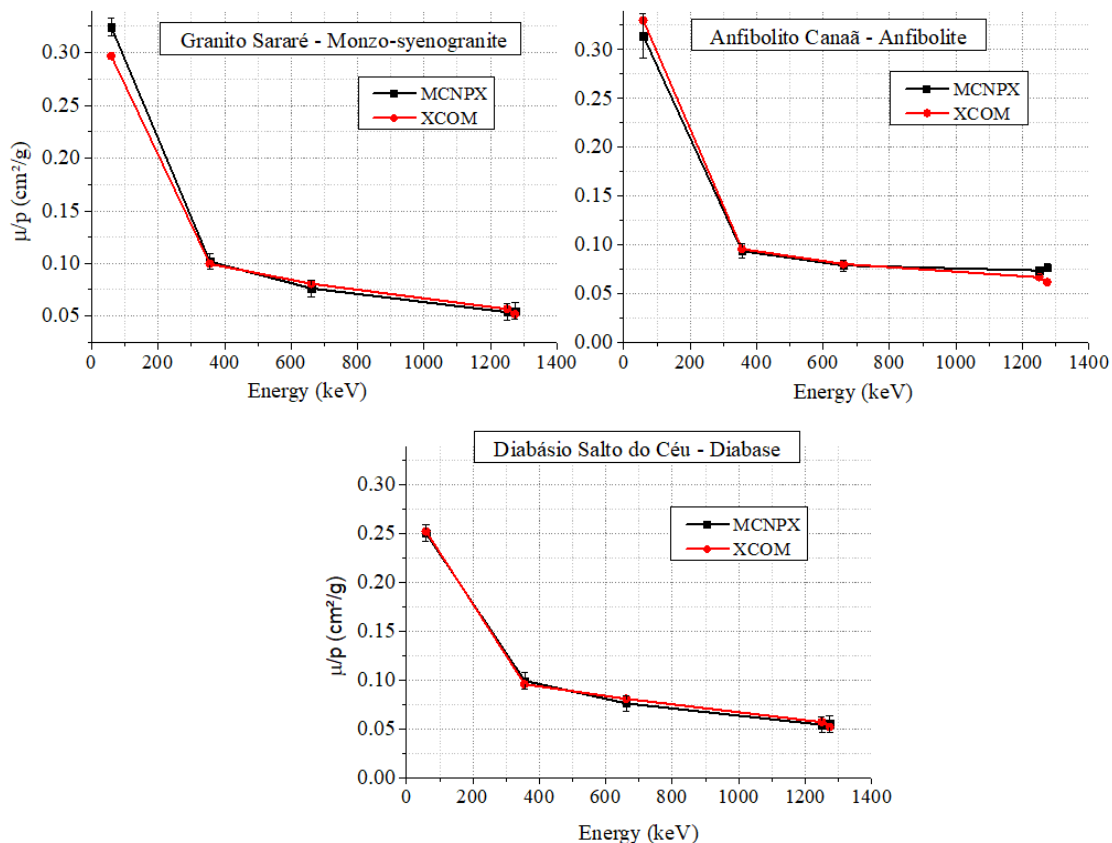


Table 2 presents the HVL as functions of the energy photons for the ten granitic rocks studied in this work. The λ results for the granitic rocks varied from 1.0 cm (*Rosa Olinda*, for photon energy 59 keV) to 4.89 cm (*Granito Sararé*, for photon energy 1274 keV), which is consistent with the obtained by Obaid et al., (2018) [8]. Table 2 shows that the lowest values for the HVL was obtained for the *Anfibolito Canaã*, and the highest values for the *Branco Savana*, which means the photon interactions happened more frequently to the *Anfibolito Canaã* and less frequently to the *Branco Savana*. The *Anfibolito Canaã* presents more promising characteristics to be used as gamma radiation shielding. It can also be seen the variation of the HVL with the photon energy, showing that in all cases it increases with the increment in the photon energy, which means that for greater values of photon energy, there is a need for a thicker material to obtain the same level of protection.

Table 2. HVL obtained using MCNPX code, for different photon energies and types of granite

Common names	Photon energy (keV)				
	59.9	356	662	1225	1274
	HVL (cm)				
Ceará State					
<i>Rosa Iracema</i>	1.03 ± 0.04	2.67 ± 0.25	3.45 ± 0.41	4.78 ± 0.79	4.85 ± 0.81
<i>Rosa Olinda</i>	1.00 ± 0.03	2.66 ± 0.25	3.44 ± 0.41	4.75 ± 0.78	4.83 ± 0.80
<i>Branco Savana</i>	1.05 ± 0.04	2.67 ± 0.25	3.46 ± 0.42	4.78 ± 0.79	4.86 ± 0.81
<i>Branco Cristal Quartzo</i>	1.03 ± 0.04	2.67 ± 0.25	3.45 ± 0.41	4.78 ± 0.78	4.86 ± 0.81
Espírito Santo State					
<i>Giallo São Francisco</i>	1.03 ± 0.04	2.65 ± 0.24	3.43 ± 0.41	4.74 ± 0.77	4.82 ± 0.79
<i>Branco Dallas</i>	1.03 ± 0.04	2.65 ± 0.25	3.44 ± 0.41	4.75 ± 0.77	4.83 ± 0.80
<i>Branco Marfim</i>	1.03 ± 0.04	2.65 ± 0.25	3.44 ± 0.41	4.75 ± 0.78	4.83 ± 0.80
Mato Grosso State					
<i>Granito Sararé</i>	1.01 ± 0.04	2.69 ± 0.25	3.51 ± 0.43	4.83 ± 0.80	4.89 ± 0.82
<i>Anfibolito Canaã</i>	0.68 ± 0.02	2.19 ± 0.16	2.92 ± 0.30	4.05 ± 0.57	4.13 ± 0.59
<i>Diabásio Salto do Céu</i>	0.97 ± 0.03	2.45 ± 0.21	3.21 ± 0.36	4.41 ± 0.67	4.50 ± 0.69

The results for HVL obtained in this work are compared to the experimental results from [8]. These results as presented in Table 3, showing a difference lower than 5%.

Table 3. Comparison between the results obtained from MCNPX simulation and from experimental studies, for the μ/ρ and HVL. $\Delta\delta$ (%) represents the difference between this study and experimental (Obaid et al., 2018) [8]

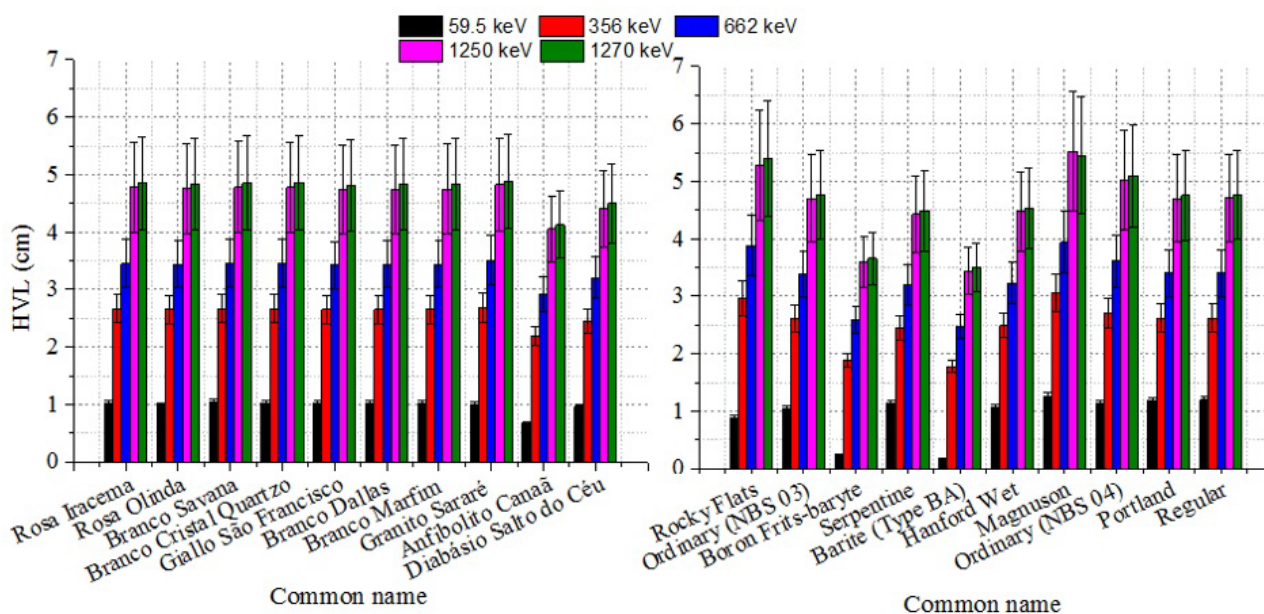
Photon energy (keV)	<i>Syenogranite - Branco Savana</i>			<i>Diabase - Diabásio Salto do Céu</i>		
	This study	(Obaid et al., 2018)	$\Delta\delta$ (%)	This study	(Obaid et al., 2018)	$\Delta\delta$ (%)
μ/ρ (cm²/g)						
122	0.152	0.167	-8.8	0.152	0.175	-13.0
511	0.084	0.089	-5.5	0.084	0.084	1.0
662	0.077	0.080	-4.2	0.080	0.077	-1.5
1170	0.056	0.060	-6.2	0.056	0.056	0.2
1275	0.055	0.057	-4.3	0.054	0.055	-1.7
1330	0.054	0.056	-2.9	0.054	0.054	-0.1
HVL (cm)						
122	1.74	1.59	9.5	1.60	1.40	13.7
511	3.15	2.98	5.6	2.87	2.90	-1.0
662	3.46	3.32	4.2	3.21	3.19	0.4
1170	4.71	4.43	6.4	4.34	4.17	4.1
1275	4.86	4.66	4.3	4.50	4.34	3.6
1330	4.88	4.74	2.9	4.51	4.47	0.9

Table3 also depicts the lack of significant differences obtained for μ/ρ and HVL when results for the granitic rocks *Syenogranite* and *Diabase* where compared with the experimental data from [8]. 13.7% was found to be the biggest difference for the HVL always for the photon energy equal to 122 keV. These differences, which are linked to the difference in the values of μ/ρ , are connected to the small variations between the densities and chemical compositions of the samples, used by Obaid et al. (2018) [8], and the values used in the present work. For a range of energy from 100 to 150 keV, the differences regarding the contribution of photoelectric effect in the total values of μ/ρ are critical, resulting in the biggest difference for the 122 keV.

The values of μ/ρ for concrete samples, which are widely used as radiation shield, are presented in Figure 5. For the calculation it was used approximately the same energy as applied in this work. Information about the density and chemical composition for all the ten types of concrete is available

on Mcconn et al., 2011 [22]. The obtained results for the HVL, to each type of concrete, were compared (Figure 5) with the results from the ten granitic rocks analyzed.

Figure 5. Comparison of the HVL for the granitic rock samples with regards to different types of concrete calculated by using MCNPX



Most part of the granitic rocks presented similar results (slightly larger) for the HVL comparing to the values obtained for the different types of concrete. However, it is important to mention that the *Anfibiolito Canaã* and *Diabásio Salto do Céu* presented the lowest values for the HVL among the analyzed compositions, which implies a superior capability to act as gamma radiation shielding in comparison to the other granitic rocks.

4. CONCLUSION

Based on the presented results, this work concludes that the granitic rocks, specially the *Anfibiolito Canaã* and *Diabásio Salto do Céu*, are promising materials to be used in radiation shield. This conclusion was taken from the analysis of their HVL values, comparing with different types of concretes. The low HVL values, in comparison with other samples, for the quoted granitic rocks, express this shield efficiency. The obtained results from the simulations are in good agreement with the experimental data extracted from the literature. It is also possible to conclude

that the geometric model built using MCNPX is an alternative method of evaluation with regards to study gamma radiation shielding properties of new types of materials, which is an important, and always demanding, area in the nuclear sciences and medical applications.

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