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A STUDY OF SELF-ATTENUATION CORRECTION FOR GEOLOGICAL MEASURES OF PARANÁ STATE GRANITES WITH HIGH RESOLUTION GAMMA-RAY SPECTROMETRY

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ABSTRACT

In high resolution gamma-ray spectrometry correct determination of ^{226}Ra, ^{232}Th and ^{40}K activities concentrations involve, beside accurate areas determinations, the use of precise efficiency calibration curves.
As the efficiency calibration curve used for activities calculations was obtained with an aqueous standard multi-radionuclides solution and the geological samples have apparent densities higher than the efficiency standard one, a correction of the efficiency curve is necessary. In this work, the self-attenuation correction factors were measured for sixteen geological samples from the Paraná State Brazil crystalline shield, including lithotypes like rhyolite, granite, sienite and basalt, with apparent densities varying from 1.42 g cm^{-3} to 2.02 g cm^{-3}. The self-attenuation factors were determined by the transmission technique and a high-resolution gamma-ray spectrometry facility, using ^{60}Co, ^{133}Ba, ^{137}Cs and ^{152}Eu punctual sources with well-known energies. For each density, a curve is fitted, allowing to correct the efficiency of the gamma transitions used in the determination of the ^{226}Ra, ^{232}Th and ^{40}K activities concentrations.

1. INTRODUCTION

Natural radioactivity is present in almost everything, like drinking water, foodstuff, soil, rocks, etc., so, the correct activities determination is a very important factor in the process of assessing effective doses and radiological indexes, in order to prevent eventual radiological impacts to humans. High resolution gamma spectrometry is the main technique in almost all radioactivity measurement laboratories worldwide as is a non-destructive, multi-elements analysis with no need of any chemical separation process and is applicable to all types of samples. Correct activities determinations involve, beside accurate areas determinations, the use of precise efficiency calibration curves. Here the challenge is when the aim of the work is to measure samples with different matrix and/or different chemical or mineralogical composition that the calibration sample. Particular energies of gamma transitions have specific penetrating habilities and, if the "real" samples and the efficiency calibration one have different matrix or chemical composition, the interaction may be different and the activity may be different of the true activity [1, 2, 3, 4]. Therefore, an accurate activity calculus must consider the self-attenuation factors together with the efficiency curve [5]. In a previous work [6], sixteen geological samples from the Paraná State Brazil crystalline shield, including lithotypes like rhyolite, granite, sienite and basalt, were measured for ^{40}K, ^{226}Ra and ^{232}Th activities concentrations determination. All samples presented apparent densities

varying from 1.42 g cm^{-3} to 2.02 g cm^{-3} and, as the efficiency calibration curve used was obtained with an aqueous standard multi-radionuclides solution in the same sample geometry, covering an energy range from 40 keV to 1600 keV, it was necessary to assess the self-attenuation factors for each sample, for the gamma transitions used in the activities determinations. The aim of this work is to calculate the self-attenuation correction factors for the sixteen samples through the transmission technique.

2. MATERIAL AND METHOD

2.1 Gamma-Ray Transmission method

The transmission technique consist basically of measuring the transmission of gamma-ray through both the geological sample and an ultrapure water sample in the same geometry, using punctual sources of Table.1, with gamma transitions in the range of interest [2, 4]. In this study were used four punctual IAEA gamma-ray sources of ^{152}Eu, ^{133}Ba, ^{60}Co and ^{137}Cs, with sixteen energies ranging from 80 keV to 1408 keV, the transmissions measurements were made for each geological sample.

Table 1. Gamma ray transitions selected for the self-attenuation study.

IAEA Calibrated Gamma-ray source [7]	Energy (keV) [8]	Intensity (%)
^{152}Eu	121.78	28.37
	244.69	7.53
	344.28	26.57
	778.90	12.97
	867.39	4.21
	1085.84	10.13
	1112.08	13.54
	1408.02	20.85
^{133}Ba	80.99	34.11
	276.39	7.15
	302.85	18.30
	356.01	61.94
	383.85	8.90
^{60}Co	1173	99.86
	1332	99.98
^{137}Cs	661	85.10

For a specific sample, a self-attenuation factor was obtained for each one of the gamma source transitions, how descript by Cutshall [2] by the equation 1,

$$f_i = \frac{\ln\left(\frac{G_i}{S_i}\right)}{\left(\frac{G_i}{S_i} - 1\right)} \quad (1)$$

Where
f_i = self-attenuation factor for a particular *i-th* gamma transition
G = beam intensity transmitted through the geological sample for a particular *i-th* gamma transition
S = beam intensity transmitted through the ultrapure water sample for a particular *i-th* gamma transition.

For each sample, a self-attenuation function was fitted, considering the attenuation factors obtained for all gamma transitions.

2.2 Gamma-ray Detection System

The gamma-ray spectra were measured by high-resolution gamma-ray spectrometry with a coaxial high-purity germanium detector (HPGe) of 15% relative efficiency and resolution of 1.9 keV for the gamma-ray transition of 1.33 MeV of ^{60}Co, with conventional electronics and an a 919 ORTEC EG&G Spectrum Master 4k-multichannel analyzer. All spectra were analyzed with the InterWinner 6.0 software [9].

2.2 Samples

The sixteen geological samples studied from the Paraná State Brazil crystalline shield, including lithotypes like rhyolite, granite, sienite and basalt, are shown in Table.2.

Fifteen samples were pulverized to 60 Mesh and sealed each one in a 100 mL HDPE flat-bottom cylindrical flask with screw cap and bubble spigot. The apparent densities varied from 1.57 g cm^{-3} to 2.02 g cm^{-3}.

In order to verify the grain size influence on the attenuation factor, one sample (Verde Tunas) was sieved in three different grain sizes, 1 mm, 2 mm and 5 mm and sealed in the 100 mL HDPE flasks, with apparent densities of 1.81 g cm^{-3}, 1.42 g cm^{-3} and 1.44 g cm^{-3}, respectively. As expected, the smaller grain size, present the higher density, as it is possible to fit more quantity in the same flask volume.

Table 2 – Geological samples from the Paraná State Brazil studied in this work.

ID	Type	Trade name	Apparent density (g cm^{-3})
*VT1MM	Sienite	Verde Tunas 1mm	1.81
*VT2MM	Sienite	Verde Tunas 2mm	1.42
*VT5MM	Sienite	Verde Tunas 5mm	1.44
RCB1	Rhyolite	Riolito Castro Brita	1.57
VI	Granite	Vermelho Itaipu	1.65
VB	Sienite	Verde Boreal	1.66
JI	Granite	Jade Imperial	1.67
VM	Sienite	Verde Mar	1.68
BD	Granite	Bege Dunas	1.69
RC	Granite	Rosa Curitiba	1.74
BP	Granite	Branco Paraná	1.75
PG	Granite	Paraná Green	1.77
CA	Granite	Cerro Azul	1.78
SFG	Sienite	Sea Foam Green	1.78
VD	Granite	Verde Duna	1.84
VV	Granite	Verde Vulcano	1.89
CI	Granite	Café Imperial	1.99
BB1	Basalt	Basalto Brita	2.02

* Samples measured for grain size influence study

3. RESULTS

3.1 Self-attenuation factors

For each studied sample, a total of 16 self-attenuation factors were obtained for all the measured gamma transitions, using Eq. 1. The results were fitted and a self-attenuation factor curve vs. gamma energy was obtained for every geological sample.
Typical fitted self-attenuation curves are shown in Figs. 1 and 2 for Granite sample Verde Vulcano (VV), apparent density 1.89 g cm^{-3}, and Granite sample Bege Dunas (BD), apparent density 1.69 g cm^{-3}.
In Figs. 3, 4 and 5 it can be seen the typical variation of the self-attenuation factors with the sample density for some energies.

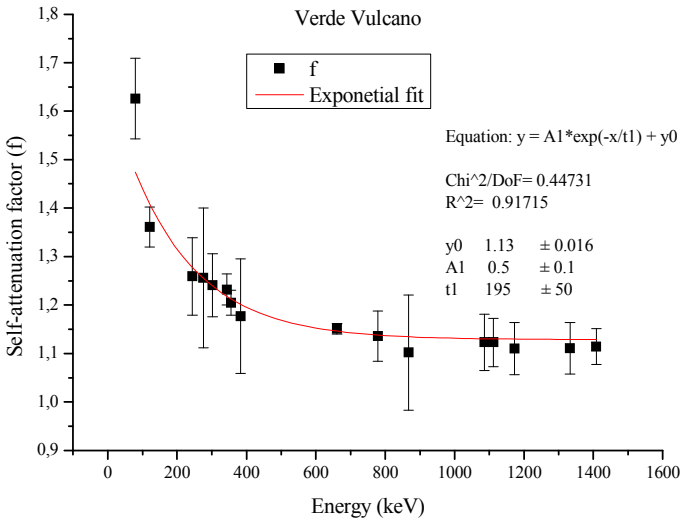

Figure 1. Self-attenuation factors obtained for the granite sample Verde Vulcano (VV).

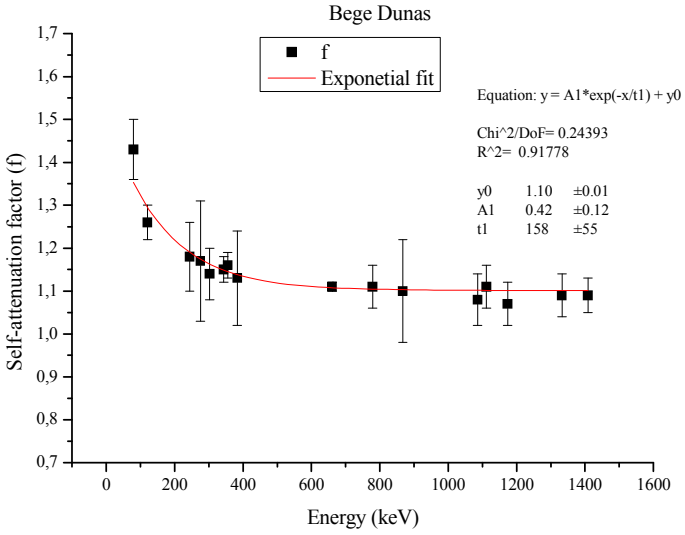

Figure 2. Self-attenuation factors obtained for granite sample Bege Dunas (BD).

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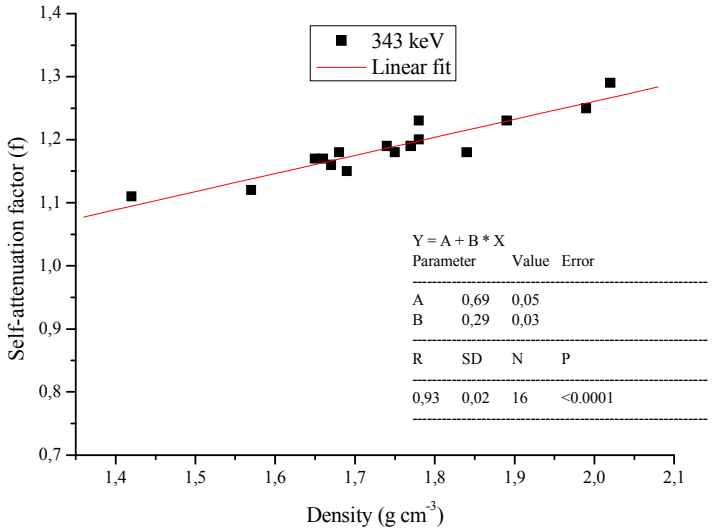

Figure 3. Self-attenuation correction factors versus density for 343 keV.

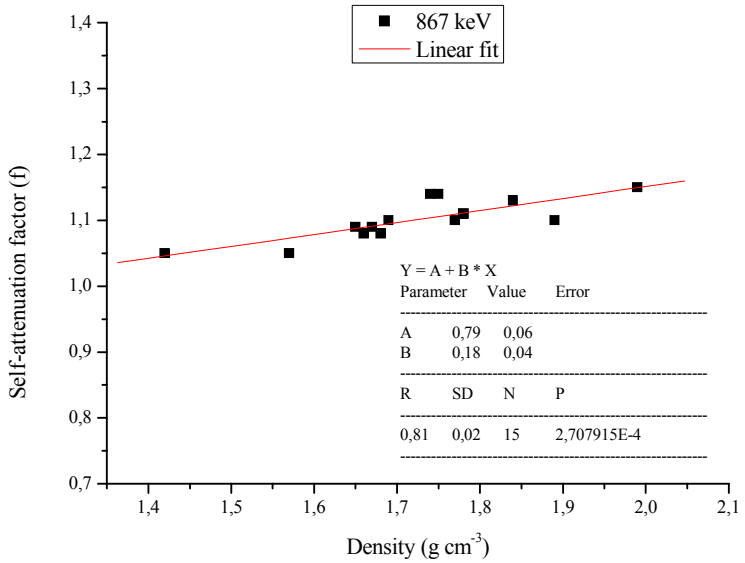

Figure 4. Self-attenuation correction factors versus density for 867 keV.

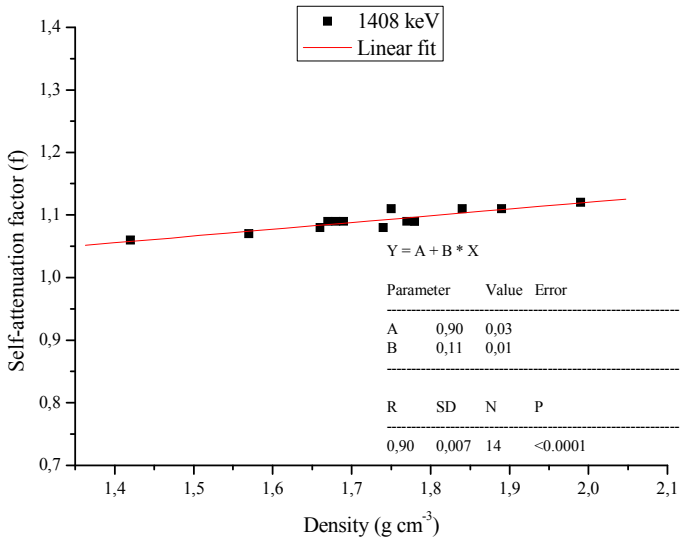

Figure 5. Self-attenuation correction factors versus density for 1408 keV.

This results show, how expected, that the self-attenuation factor increases with the sample apparent density and decreases with the gamma transition energy.

For all samples, the mean increase in the activities due to the self-attenuation correction will be around 10% for ^{40}K, 16% for ^{226}Ra and 19% for ^{232}Th.

3.2 Grain size influence study

For a particular sample, three different grain sizes were analyzed and the results showed in Fig. 6.

The higher attenuation factors were found for the higher density, as showed also in Figs. 3, 4 and 5.

The attenuation factors for the 2 mm and 5 mm show quite the same values, as the samples apparent densities are almost the same.

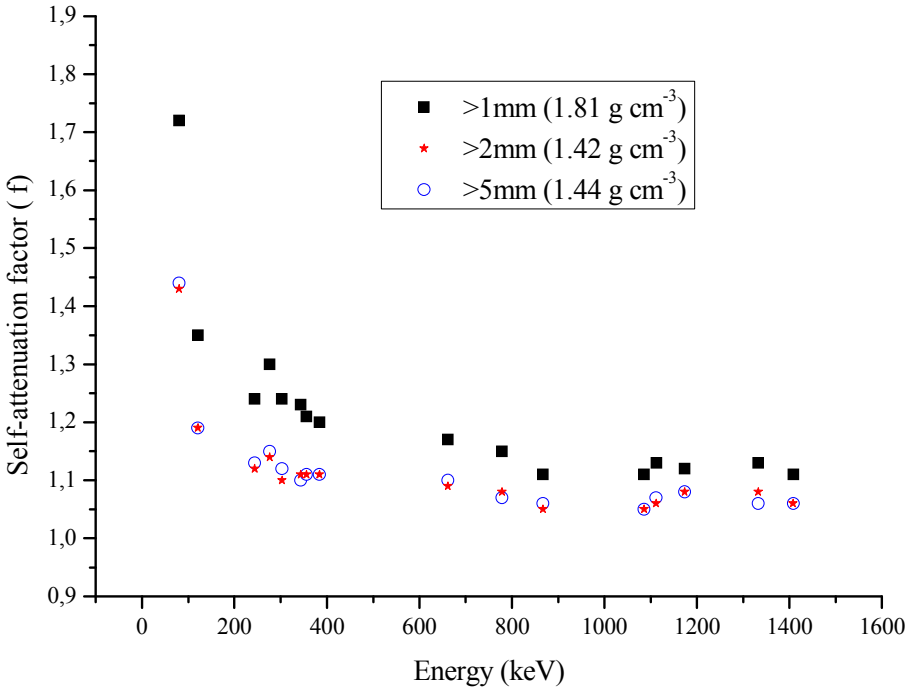

Figure 3. Self-attenuation factor as function of sample grain size

3. CONCLUSIONS

The results showed that the apparent density has a strong influence on the self-attenuation factor, even for medium gamma energies, and any activity concentration result must consider the sample self-attenuation.

By now, it seems that the grain size has a minor influence on the sample attenuation, but, surely, more studies are necessary.

The relationship between the mineral composition of geological samples and the samples self-attenuation will be the subject of further studies.

ACKNOWLEDGMENTS

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