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IRON INFLUENCE IN SELF-ATTENUATION OF GAMMA RADIATION IN SAND SAMPLES

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ABSTRACT

Self-attenuation of gamma rays are usually observed in sand samples, due to the presence of iron particles of natural or anthropogenic causes, which may lead to misleading activities concentrations results. As iron atoms are often present in sand samples, the self-attenuation contribution from this mineral requires special attention when assessing sand samples radioactivity. This contribution is verified in the present study, using several silica (sand-like) samples doped with different concentrations of iron. The samples were arranged with pure SiO4 (98,9%) mineral from the region of Setiba, (Espírito Santo state, Brazil) and Itabirito mineral, from Itabira (Minas Gerais state, Brazil), using standard 100 mL high density polyethylene (HDPE) flat-bottom cylindrical flasks, each sealed using a 52.5 mm plan screw cap and bubble spigot. Three samples were arranged in different concentrations – Fe 25% and SiO_4 75%; Fe 50% and SiO_4 50%; Fe 75% and SiO_4 25% and two other samples were assembled in the same geometry, using 100% SiO_4 and 100% Fe. These samples were exposed to a large spectrum of gamma radiation (121 keV to 1408 keV) emitted by sources of ^{60}Co, ^{137}Cs and ^{152}Eu. These gamma radiations were collimated and directed over the doped samples in a high resolution gamma ray spectrometer, whose spectra were analyzed with the WinnerGamma software. The matrix effect intensity is inferred by the percentage differences observed in varying the iron concentration that proved to be more intense for lower energy radiation.

1. INTRODUCTION

Environmental radiation measurements involve a series of techniques that can estimate the type of radioactive nucleus present in a sample and its activity. Depending on the density and composition of the sample being analyzed, the radiation emitted on its radioactivity decay can be partially absorbed by the sample and hence, not be collected by the detector. This effect is known as self-attenuation gamma, when the decay of the sample involves gamma rays [1].

Interactions of gamma rays in a particular material can promote absorption of energy and scatter radiation, mainly due to photoelectric effect, Compton effect and pair production, and the primarily effect will depend mostly on the incident radiation energy and on the atomic number of the target material. When measuring radiation that undergo such effects in the target material, the radiation that is either absorbed or scattered in a sample promote a matrix effect that, if not properly corrected, can result in misleading measurements.

In a previous study using gamma ray spectrometer to analyze sand samples, the matrix effect yielded by absorption or scattering in the sample (in other words, the self-attenuation effect)

was observed [2]. Ordinary sand is a composition of different kinds of minerals (depending on the type of rocks found locally) in which silica is the most common one [3]. Also, other minerals can be normally found within sand formations, including iron particles.

Independently of sand densities, the referred study [2] has shown that the presence of iron particles, of natural or anthropogenic cause, has contributed for the self attenuation effect. Also, this effect is more prominent for lower energies (from a few keV up to 600 keV), where the photoelectric effect and Compton scattering are more probable to take place for materials with atomic number about 20 (most common rocks) [4]. Once iron particles may take part in the composition of sands, it is interesting to infer its contribution for the self attenuation effect, when studying these types of samples.

In the present study, the self attenuation effect was calculated in samples of combined compounds – silica and iron, with known concentrations. The calculation will later allow inferring the self attenuation contribution of iron ions in sand-like type of samples, minimizing misleading measurements that could occur when studying samples of such type.

2. MATERIALS AND METHODOLOGY

2.1. Sample Setup and Measurements

Five sand-like samples were designed to simulate ordinary sand samples, combining known concentrations of silica (SiO_4 mineral, from the region of Setiba – Espirito Santo state, Brazil) and iron powder (Itabirito mineral, from Itabira – Minas Gerais state, Brazil). Pure silica was chosen to compose the designed samples not only due to its large presence in ordinary sand [3], but also to assure (from its purity) that no iron ions would be present. The purity of the SiO_4 was verified using wavelength dispersive X-ray fluorescence method (WDXRF), whose results shown 98,9% of SiO_4.

Three samples were arranged in different concentrations – Fe 25% and SiO_4 75%; Fe 50% and SiO_4 50%; Fe 75% and SiO_4 25% using standard 100 mL high density polyethylene (HDPE) flat-bottom cylindrical flasks, each sealed using a 52.5 mm plan screw cap and bubble spigot. Also, two other samples were assembled, in the same geometry, using 100% SiO_4 and 100% Fe.

All samples were irradiated using a punctual standard radioactive source (IAEA-EMS), placed over a 2 mm open lead collimator with 10 mm thickness, that was positioned over the sample. The collimator-sample setup was then positioned over a coaxial high-purity germanium detector (HPGe), as shown in Fig. 1, with an outermost lead shield of 10 cm thickness.

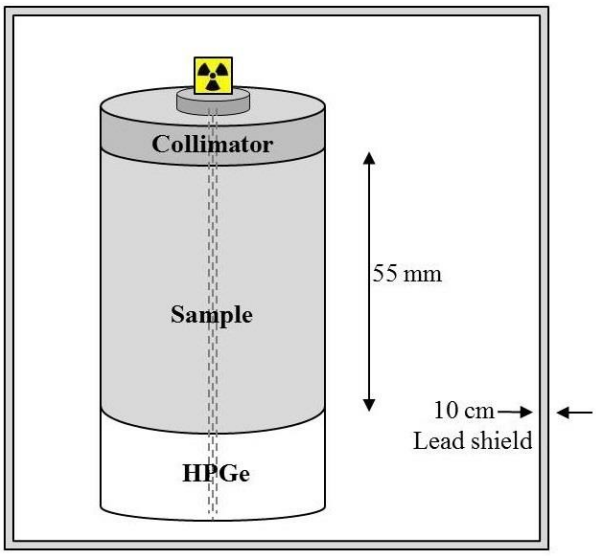

Figure 1: Merely illustrative scheme of the experimental setup.

The HPGe detector presents 15% relative efficiency with conventional electronics and a 919 ORTEC EG&G Spectrum Master 4k multichannel analyzer. The measured resolution for the ^{60}Co 1332.5 keV is 2.9 keV. Spectra were analyzed with the WinnerGamma software [5-6].

All nuclides activities are given with uncertainty statistics at ±1σ confidence level and the detections limits are given at ±2σ confidence level with the GTN5 formulae. The detector efficiency curve was determined with a multi-radionuclide standard aqueous solution in the same geometry as all measured samples.

2.2 Percentage of Iron Influence

Iron concentration within the samples can cause the matrix effect. The percentage of iron influence (P_{IF}) is calculated using the relative percentage between measurements of the 0% Fe (100% SiO4) sample and the measurements of the 25% Fe, 50% Fe and 75% Fe samples:

$$P_{IF} = \frac{(A - A_d)}{A} \times 100\% \qquad (1)$$

Where A represents the liquid areas for peaks in the 0% Fe sample and A_d represents the liquid areas for peaks in iron doped samples (25% Fe, 50% Fe and 75% Fe). The uncertainty can be obtained applying the relative standard deviation.

3. RESULTS AND DISCUSSIONS

Every sample was exposed to the radiation emitted by ^{137}Cs, ^{152}Eu and ^{60}Co sources and the most intense gamma emissions were considered and compared to the ones collected for the sample with no additional iron concentration (0% iron concentration). Using equation 1 one could infer the percentage of iron influence, as shown in Table 1 and Fig. 2.

Table 1: Percentage difference calculated for different sand samples, doped with different concentrations or iron.

Element	Energy* [keV]	γ-Ray intensity* [%]	Percentage of iron in sample			
			25%	50%	75%	100%
			P_{IF}**			
^{152}Eu	121.6	28.0	23%	28%	30%	39%
	344.2	26.5	9%	9%	9%	9%
^{137}Cs	661.7	85.1	3%	2%	0%	1%
^{152}Eu	778.9	12.9	5%	5%	0%	3%
	964.2	14.6	8%	6%	6%	6%
	1112.0	13.6	7%	9%	6%	7%
^{60}Co	1173.2	99.9	2%	3%	2%	6%
	1332.5	99.9	4%	4%	3%	9%
^{152}Eu	1407.9	21.0	1%	2%	3%	2%

* [7]

** Measurements with 68% (± 1σ) confidence level, k=1 and the results present a relative uncertainty value of 2%.

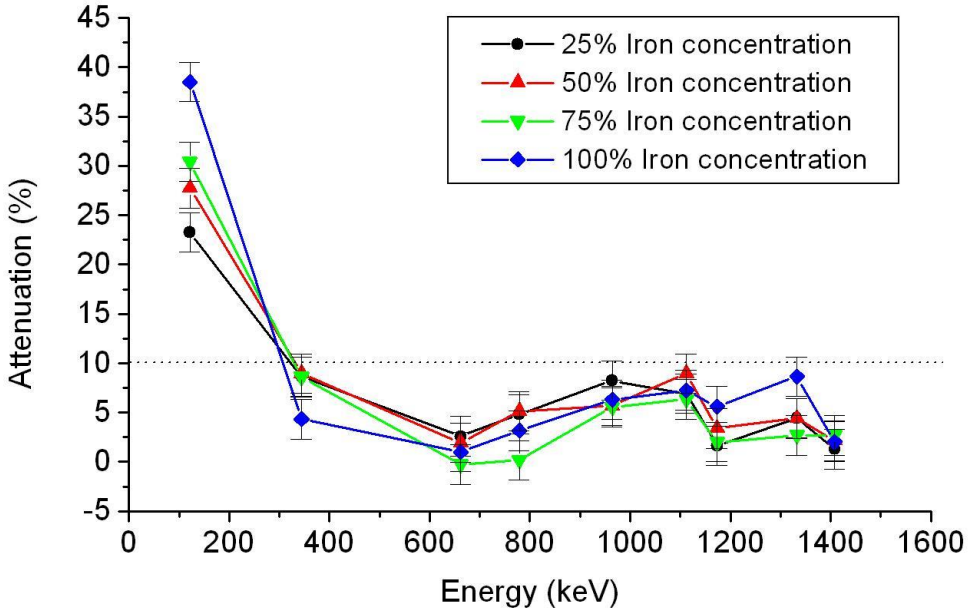

Figure 2: Percentage of iron influence observed in iron doped samples

Table 1 and Fig. 2 show that the attenuation percentage is more severe for lower gamma energies (up to about 300 keV), as expected, once the cross section of gamma rays in the atom is higher for lower energy photons [1] (considering both photoelectric absorption and Compton scattering) [8]. Therefore, when studying radionuclides that decays through emission of such energies, one must certainly use attenuation correction factors, that may be as high as 30 or 40%.

For high energy gammas, Fig. 2 shows that no attenuation correction would be necessary, curves follow a statistical fluctuation about a constant value (no higher from 10%), showing that the influence of iron is of little significance. This behavior is also expected, as the cross section of high energy gamma rays around 1400 keV tends to lower values [1].

Empirically, the curves seen in Fig. 2 and the results suggests an equation for the percentage influence $P_{IF}(E)$ as a function of energy such as:

$$P_{IF}(E) = c_1 E^{-C_2} \tag{2}$$

Where c_1 and c_2 are adjustable constants and ε is energy in keV. In the case of high energies photons, the function presents a maximum value of 10% and no correction is necessary. Considering that P_{IF} is a reduction percentage value, is possible to make an inferential correction, following equation 3

$$C_{ICF} = C \cdot \left(1 + \frac{P_{IF}}{100\%}\right) \pm \delta C_{ICF} \tag{3}$$

Where C_{ICF} is the activity concentration in $Bqkg^{-1}$, C is an attenuated activity concentration in $Bqkg^{-1}$, P_{IF} the percentage influence and δC_{ICF} the uncertainty for the activity concentration. The uncertainty can be obtained applying the expanded error expression:

$$\delta C_{ICF} = C_{ICF} \sqrt{\left(\frac{\delta P_{IF}}{P_{IF}}\right)^2 + \left(\frac{\delta C}{C}\right)^2} \tag{4}$$

The attenuation on the total counts was observed for low and high energy gamma rays as shown in Fig. 3 – 8. These are the spectra obtained before their analysis using the WinnerGamma software, therefore, they represent the gross count obtained.

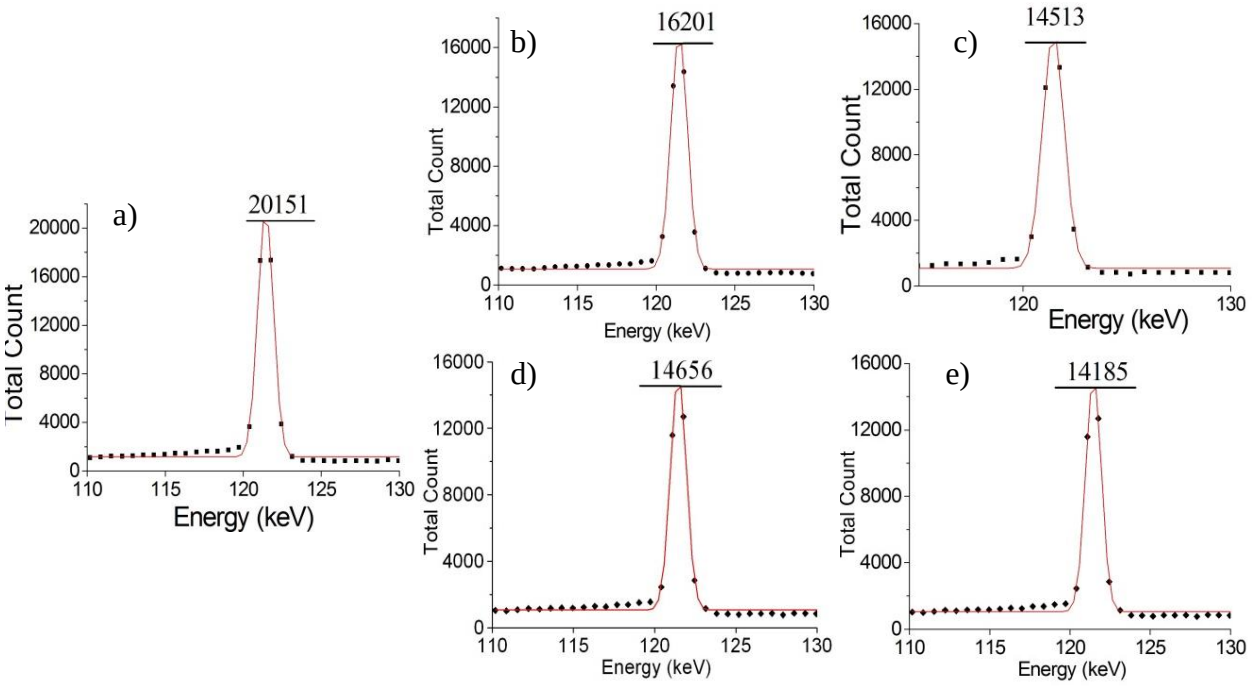

Figure 3: Total count attenuation for samples doped with 0% iron (a), 25% (b), 50% (c), 75% (d) and 100% (e), measured for the gamma energy of 121.6 keV from ^{152}Eu.

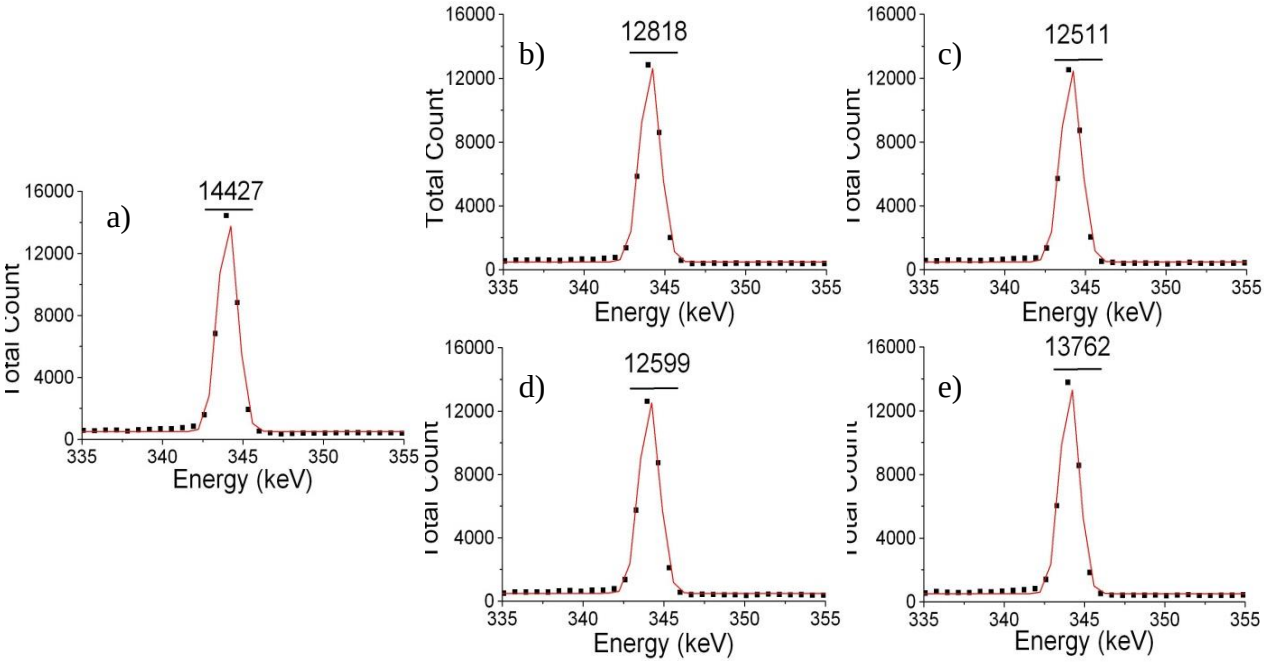

Figure 4: Total count attenuation for samples doped with 0% iron (a), 25% (b), 50% (c), 75% (d) and 100% (e), measured for the gamma energy of 344.2 keV from ^{152}Eu.

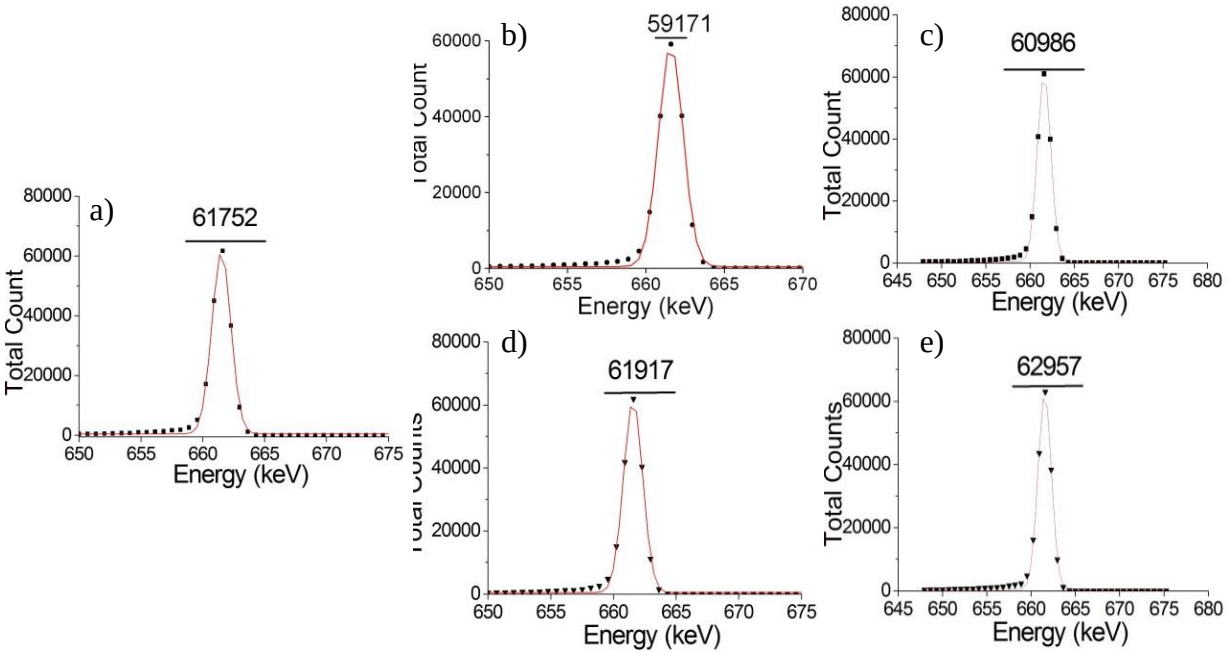

Figure 5: Total count attenuation for samples doped with 0% iron (a), 25% (b), 50% (c), 75% (d) and 100% (e), measured for the gamma energy of 661.7 keV from ^{137}Cs.

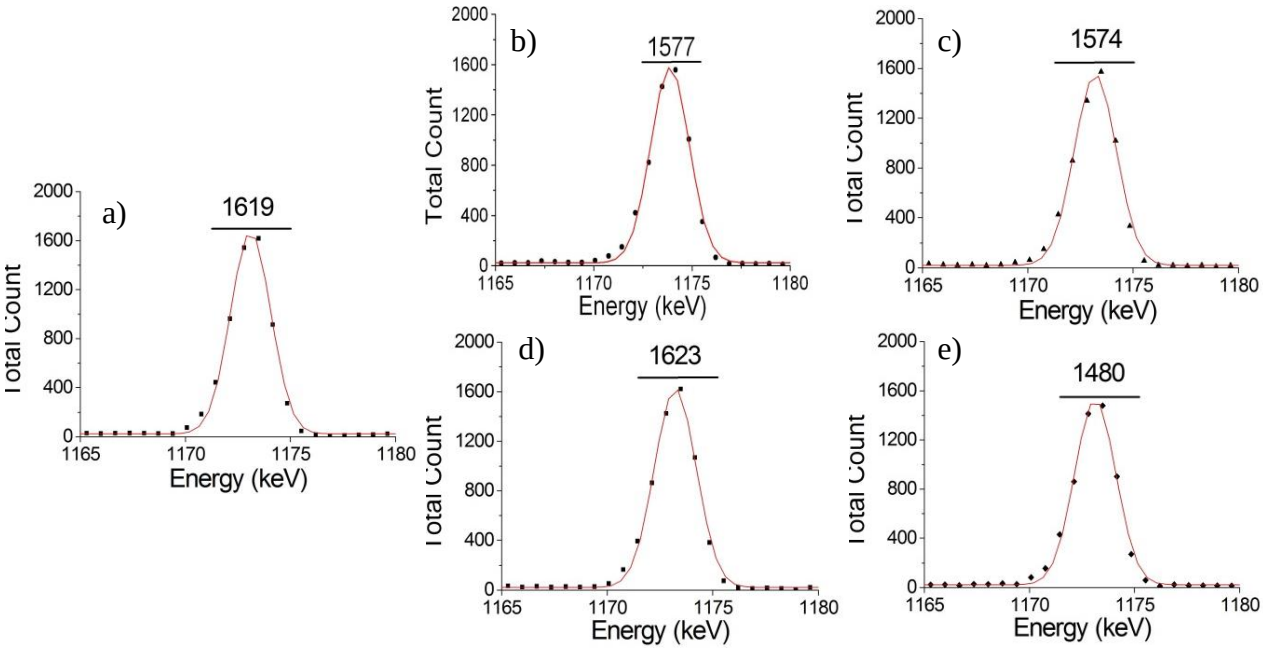

Figure 6: Total count attenuation for samples doped with 0% iron (a), 25% (b), 50% (c), 75% (d) and 100% (e), measured for the gamma energy of 1173.2 keV from ^{60}Co.

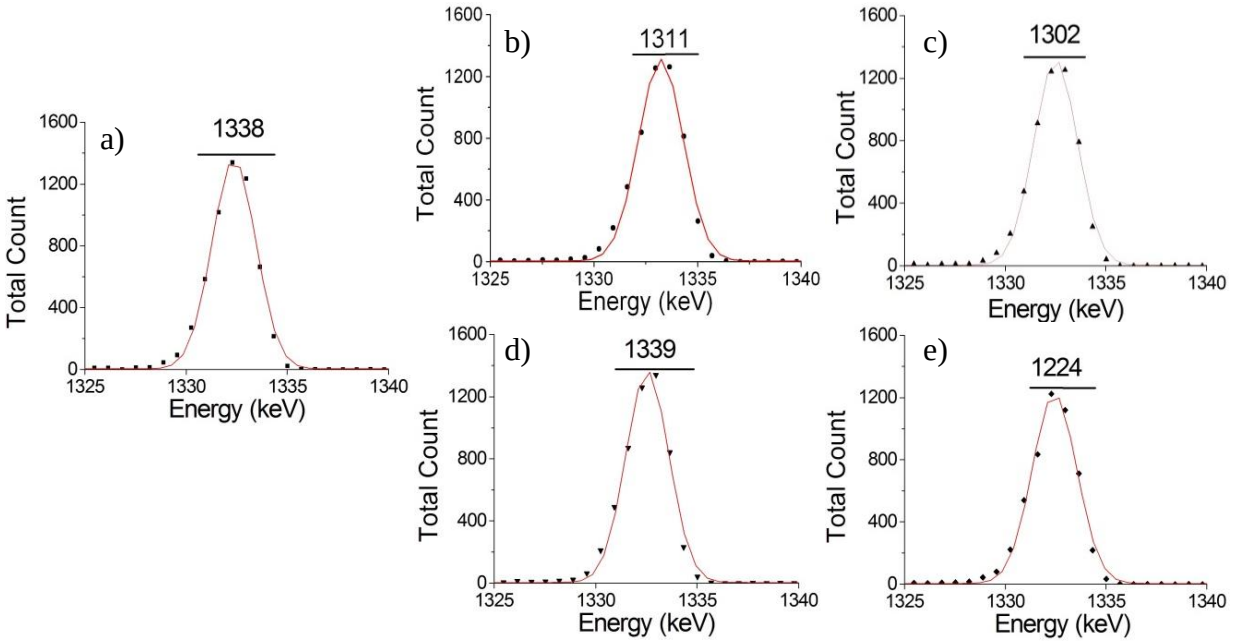

Figure 7: Total count attenuation for samples doped with 0% iron (a), 25% (b), 50% (c), 75% (d) and 100% (e), measured for the gamma energy of 1332.5 keV from ^{60}Co.

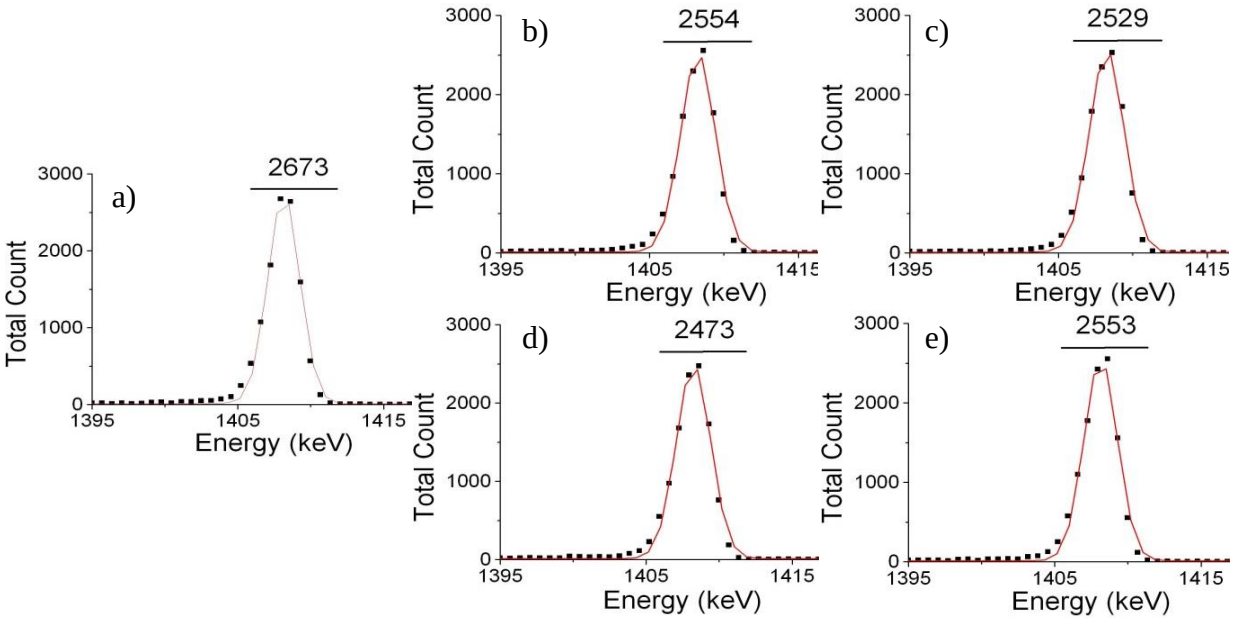

Figure 8: Total count attenuation for samples doped with 0% iron (a), 25% (b), 50% (c), 75% (d) and 100% (e), measured for the gamma energy of 1407.9 keV from ^{152}Eu.

Considering Fig. 3, the attenuation difference observed for all of the samples, within its statistical uncertainties, is sharper than the differences observed in Fig. 4-8. This behavior strengthens the expected attenuation discussed for lower energy electromagnetic radiation and emphasizes the need of correction.

The samples arranged for this study are a composition of different compounds, including iron and silica (SiO_4). Therefore, the attenuation of gamma rays is expected from their interaction with the whole mix of particles within the sample. In Fig. 3 – 8, the samples homogenized with iron and silica show a slightly higher attenuation when compared with the samples with iron alone (100% iron sample). This is observed by the difference in amplitude of the samples with iron alone. This behavior proves that, although iron particles play an important part in the attenuation process, other minerals present within the sand may also be influencing the measurements, especially for low energy photons, as discussed earlier.

These results suggest that, when studying standard sand samples, one should use correction factors for the self attenuation behavior, to obtain more reliable analyses in the lower part of the energy spectrum, while the correction factor for the higher energy part of it is practically indifferent.

4. CONCLUSIONS

The present study has shown the matrix effect yielded by photon absorption or scattering within sand-like samples. For low energy photons (up to about 300 keV), Table 1 shows that the percentage iron influence can be as high as 39%, suggesting that self attenuation correction factors must be considered when using sand type samples. When considering higher energy photons (300 keV up to 1408 keV), it was shown that the attenuation differences lie within a constant statistical fluctuation, no higher than 10%, as shown in Fig. 2, and therefore, depending on the application, does not require self attenuation correction. Whenever correction is necessary, the proposed function (equation 3) is well suited with its associated uncertainty (equation 4).

Fig. 3 – 8, corroborate the previously results from in Table 1 and Fig. 2, showing that the attenuation of gamma rays within the sample is more evident for lower energy photons and less apparent for higher energy ones, once photoelectric effect and Compton scattering are more probable to take place for materials with atomic number close to common rocks, and therefore the attenuation is more significant.

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REFERENCES

1. EVANS, R. D., *The Atomic Nucleus*, MacGraw-Hill, New York, USA(1972).

2. AQUINO, R. R. *Avaliação da Radioatividade Natural em Areias das Praias da Grande Vitória, Espírito Santo.* IPEN, Sao Paulo, 2010.
3. SPERANDIO, E.F. *Radioatividade Natural e Composição das Areias da Praia de Camburi (Vitória– ES, Brasil). Master Degree Thesis*. Universidade Federal do Espírito Santo, Vitória, 2011.
4. IAEA – International Atomic Energy Agency. *Guidelines for radioelement mapping using gamma ray spectrometry data*. Technical Report 1363, Viena, 2003.
5. ORTEC, InterWinnerTM6.0 MCA Emulation, Data Acquisition, and Analysis Software For Gama and Alfa Spectroscopy, TOMCOM Software Ltd., 2004.
6. KNOLL, Glenn F., *Radiation Detection and Measurement*, John Wiley & Sons, New York, NY, USA. (1999). 3rd ed.
7. NNDC - National Nuclear Data Center, **Brookhaven National Laboratory**, base in ENSDF, Nuclear Wallet Cards (1996-2003). Available: <http://www.nndc.jbl.gov>, access: 26/07/2013
8. PODGORSAK, Ervin B. *Radiation Physics for Medical Physicists*. Springer, London, New York, USA. (2009). 2rd ed.

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