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High density concrete to application in radiological protection. Asesement of the additions-microsilica-metakaolin-limestone filler.

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ABSTRACT

Currently, the use of additives in the production of high density concrete for gamma and X radiation protection, have happened more frequently than ever. The additives contribute to the reduction of cement consumption and CO_2 emission, and also improve the performance in the application and usage, as well as increment the linear attenuation coefficient. In Brazil are used mainly the addition of microsilica, metakaolin and limestone filler. In order to understand the contribution in performance of the concrete for radiation protection given these additives, this paper evaluates the effects of additives in function of the chemical elements that are present in raw materials that were utilized, so that the draftsman can choose the raw material to be applied in the high density concrete, in function of the quantity and type of element that is present. To conduct those experiments, test bodies of concrete were produced by applying a specific methodology that controls parameters such as: dosage, water cement ratio, vibration energy, vibration time and maturity. The concretes utilized had as basic structure of raw materials such as ciment, magnetite sand plus separate portions of metakaolin, microsilicum and limestone filer. In order to gather the data of gamma transmission, a computerized micrometric table was used, automated with a controlled displacement system in x and y axes, a 137Cs source with 3.7 10^{10} Bq of activity and energy E = 660 keV, output collimator of source in lead with opening of 2mm and input collimator detector with the same characteristics; An Ortec 3”x3” detector, NaI (Tl) sodium iodide activated with thallium, with standard electronics composed by amplifier, source of tension, and multichannel with single channel functionality. The results found showed that the variations of chemical elements in the sampled concretes provided significant alterations and different results.

1. INTRODUCTION

Utilizing additions to high density concretes in order to improve shielding against gamma and X radiations is an option that offers some advantages,

It contributes to improving the absorption coefficient, decreases the consumption of cement by replacing the same, therefore reducing CO_2 emissions that results from cement manufacture.

In Brazil, the concrete additions are carried out with the utilization of metakaolin, microsilica and limestone filler as materials.

Differing in compositions and grain sizes, the effects produced in the concretes used for the structures are similar, while for high density concretes meant for protecting against gamma and X radiations, the performance when used for shielding, when proportions are kept, differs due to the prevailing effect between radiation and the matter, which depends on the radiation energy to be reduced.

The chemical compositions of the constituent materials mentioned differ especially in their contents of CaO and SiO_2.

In the energy band where photoelectric effects prevail, it is important that the atomic number is elevated and the cross section is a direct function of the 4th power of the atomic number and an inverse function of the 3rd power of the photon energy, while for a higher energy band where the Compton effect prevails, the important is the density of the element or compound, since the Compton effect is directly proportional to the electronic density of the material.

The utilization of additions such as metakaolin, microsilica or limestone filler in high density concrete can meet the two needs.

If those are chosen based on the chemical composition with the largest cross section for the prevailing effect, they will improve the absorption coefficient and diminish the thickness needed for shielding and the consumption of raw materials.

2 **Methods and Results.**

2.1 Methodology

With the aim of understanding and assessing the influence of additions to high density concretes, three families of microconcrete have been prepared by utilizing as aggregate the magnetite and Portland cement CP V ARI and three types of additions: microsilica, metakaolin and limestone filler.

For preparing the concretes of each family, the content of water cement ratio and aggregate cement ratio and the contents of the additions were changed, which were of 3%, 5% and 10% in relation to the Portland cement.

2.2 Raw materials for the assayed concretes.

The used matrix for developing the studies was based in the utilization of a fine aggregate of magnetite and CP V cement prepared with the above mentioned additions in a mass on the cement used and the water cement ratio was kept constant.

2.3 Physical and chemical characteristics of the raw materials.

The raw materials utilized have been both chemically and physically characterized.

The physical characteristics focused on verifying the grain size and density.

In order to know the chemical composition of the raw materials, the X Ray Fluorescence technique has been used.

2.4 Molding of test specimens

The molding of test specimens was made through manufacturing the test specimens with the utilization of a special mould shown in figure 1, which was prepared for accommodating concrete plates with a thickness of 20 mm, length of 100 mm and height of 50 mm.
By utilizing a vibratory table with vibration time and energy control, five test specimens were molded for each concrete family having as additions the chosen percents.
Afterwards, the test specimens were cured in a closed environment with controlled humidity.

Fig. 1 – Mould for molding test specimens.

2.5 Determining the theoretical linear attenuation coefficient.

The theoretical linear attenuation coefficient was calculated based on the chemical analyses of the assayed concretes multiplied by the theoretical cross sections calculated with the help of NIST (2) database whose total cross section $\frac{{}_a\mu}{\rho}$ per atom can be calculated by the relationship:

$$ {}_a\mu / \rho = \sigma_{tot} / uA \text{ (equation 1).} $$

Where u = 1.6605402x10-24 g (atomic mass unit)
A = atomic mass number.

σ_{tot} is the total cross section for interaction with the photon, whose unit is regularly given in b/atom (barns/atom), where b = 10^{-24} cm^2

$$ \sigma_{tot} = \sigma_{ph} + \sigma_{com} + \sigma_{pair} \text{ (equation 2)} $$

where:

σ_{ph} cross section for the photoelectric effect.

σ_{com} cross section for the Compton effect.

σ_{pair} cross section for the production of pairs.

The results shown in the tables that follow refer to:

$$\mu/\rho = \sigma_{tot}/uA$$

Now, in table 1, we show the results of the theoretical linear attenuation coefficients.

Table 1 – Theoretical linear attenuation coefficients

		Specific mass g/cm^3		**Theoretical attenuation coefficient660 keV**	
Metakaolin	10%	**2,540**		0.1945	
Microsilica	10%	**2,640**		0.2019	
Metakaolin	5%	**2,820**		0.2160	
Filler	10%	**2,900**		0.2198	
Metakaolin	3%	**2,940**		0.2266	
Microsilica	5%	**3,150**		0.2382	
Microsilica	3%	**3,320**		0.2539	
Filler	3%	**3,270**		0.2502	
Filler	5%	**3,360**		0.2526	

2.6 Determining the experimental linear attenuation coefficient.

The experimental linear attenuation coefficient was measured in the facilities of the applied nuclear physics laboratory of the physics department of the *Universidade Estadual de Londrina* (Londrina State University) by using an automated gamma transmission system that allows the acquisition of points regularly spaced, composed by a 3X3" NaI (TI) detector with entrance collimation of 3 mm, source of Cesium-137 with 3.7 10^{10} Bq activity using collimated beam with 2 mm of diameter. In each sample points at each 5 mm were measured totaling seven positions divided by three horizontal lines spaced at each 10 mm.

After being cured in open air, the test specimens were assayed by gamma ray transmission with the use of the above mentioned system, with gamma rays energy of 660 keV, for determining the linear attenuation coefficient through the D'Lambert Law, shown in equation 3.

$$I = I_0 e^{-\mu x} \qquad \text{(equation 3).}$$

Where I is the intensity of the gamma rays beam transmitted by the sample, I_0 is the intensity of the gamma rays beam before crossing the sample, μ is the linear attenuation coefficient of the concrete and x is the thickness of the test specimen, which in this work was 20 mm. The measures acquisition system is shown in figure 2.

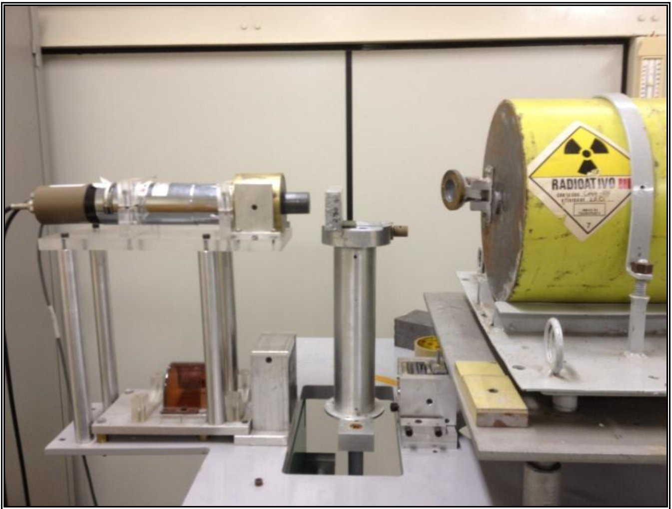

Figure 2 – Measuring table with ^{137}Cs source.

Radiations were measured in the samples in three vertical lines and seven horizontal points in each line.

Table 2 contains the experimental data that were measured, which completes table 1.

Table 2 – Theoretical linear attenuation coefficient, experimental linear attenuation coefficient and experimental cross section.

		Specific mass g/cm^3	**Linear attenuation coefficient cm^{-1}**	**Theoretical attenuation coefficient 660 keV cm^{-1}**	**Experimental cross section cm^2**
Metakaolin	10%	**2.540**	0.1815	0.1945	0.0715
Microsilica	10%	**2.640**	0.1985	0.2019	0.0752
Metakaolin	5%	**2.820**	0.2130	0.2160	0.0755
Filler	10%	**2.900**	0.2270	0.2198	0.0783
Metakaolin	3%	**2.940**	0.2320	0.2266	0.0789
Microsilica	5%	**3.150**	0.2395	0.2382	0.0760
Microsilica	3%	**3.320**	0.2570	0.2539	0.0774
Filler	3%	**3.270**	0.2570	0.2502	0.0786
Filler	5%	**3.360**	0.2650	0.2526	0.0789

2.7 Relationship between the linear attenuation coefficient and the energy absorption coefficient.

As we utilized the Cesium-137 gamma radiation, there was no production of pairs and therefore, equation (1) is reduced to two terms after the equal sign.
As it is the case with Compton spreading, the interaction gamma ray is spread with a lower energy and, therefore, it is not totally absorbed in the equation (4) and we shall introduce a Build-Up factor, and consequently, equation (5) changes to

$$I' = I_{0.}\, B.\ e^{-\mu x} \qquad \text{(equation 4), where}$$

B is the Build-Up factor.
This equation can be written by utilizing the μ_{en} energy absorption coefficients, and then equation (3) becomes

$$I' = I_{0.}\, B.\ e^{-\mu x} = I_{0.}\ e^{-\mu\ en\ x} \qquad \text{(equation 5).}$$

2.8 Conclusion.

This work aims at assessing which addition is the most suitable for shielding against gamma and X radiations with energy of 660 keV.
Figure 3 shows the results, where we can observe that the addition of 5% of limestone filler was the one that showed the largest linear attenuation coefficient, followed by the additions of 3% of limestone filler and 3% of microsilica.

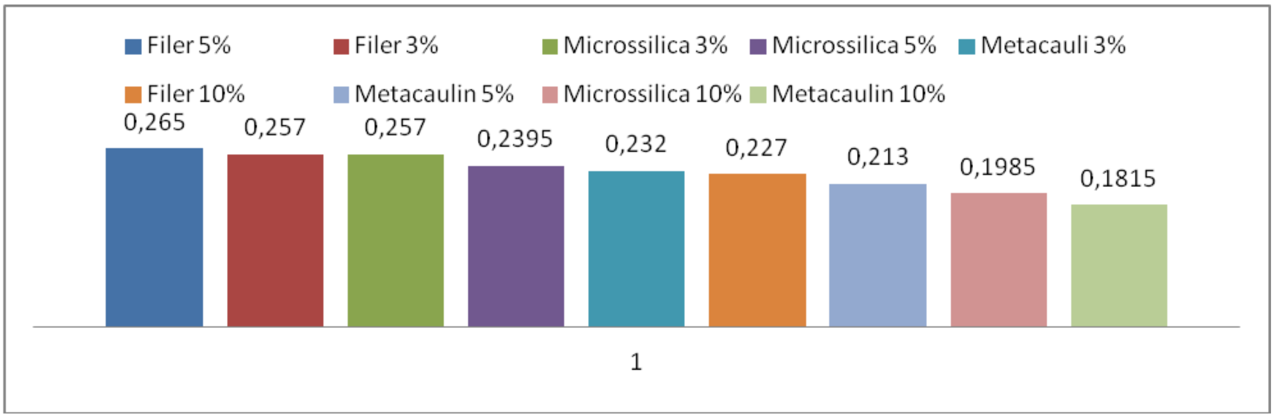

Figure 3 – Coefficient chart of experimental linear attenuations for the different types of concretes with additions.

It is possible to observe in table 2 previously shown, a little difference between the linear attenuation coefficients, which were measured and calculated. Those differences there is due to the fact that by the calculation of the theoretical linear attenuation coefficient, a percent combination of each constituent chemical element is assumed without taking into consideration their grain size and space existing in their molecular structure, while what was experimentally determined shows the reality of the composition by taking into account those factors.

The fact that the quantities of light elements existing in the compounds have not been surveyed is also a factor contributing to justify the difference found between the theoretical linear attenuation coefficient and the experimental linear attenuation coefficient.

It is important to point out that the utilization of chemical analysis to find the theoretical linear attenuation coefficient is a practice that can help the designer to choose the components to formulate a concrete trait to be used for shielding against gamma and X radiations, transforming the result found into a effective atomic number in order to compare it with that chemical element.

If we take into account that the free middle way between two successive collisions with the shielding medium is given by equation $1/\mu$, the results of the free middle ways for the compositions of the materials used can provide the following results shown in table 3 beneath.

Table 3 – Free middle way for the experimental attenuation coefficients

Concrete types	**Additions (%)**	**Linear attenuation coefficient (mcm^{-1})**	**Free middle way (cm)**
Metakaolin	10%	0.1815	5.5096
Microsilica	10%	0.1985	5.0377
Metakaolin	5%	0.2130	4.6948
Filler	10%	0.2270	4.4052
Metakaolin	3%	0.2320	4.3103
Microsilica	5%	0.2395	4.1753
Microsilica	3%	0.2570	3.8910
Filler	3%	0.2570	3.8910
Filler	5%	0.2650	3.7735

As the thickness of our sample is of two centimeter, it can be concluded that the experiment shows a narrow beam and condition of good geometry, i.e. build-up factor is equal to one, and therefore equation 3 is valid.

Similarity of the cross section

For concretes with additions of 3%, 5%, 10% of limestone filler and 3% of metakaolin, the cross section of the compounds is similar to Calcium, whose Z = 20 and the cross section for 660 keV is approximately 0.0784 cm^2.

For the other concretes we can attribute the behavior to the Potassium element with Z = 19, with the exception of the concrete with 10% addition of metakaolin, whose behavior can be compared to the Argon with Z = 18.

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