2011 International Nuclear Atlantic Conference - INAC 2011
Belo Horizonte,MG, Brazil, October 24-28, 2011
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: **978-85-99141-04-5**

STUDY OF THE RELATIONSHIP BETWEEN PEAKS SCATTERING RAYLEIGH TO COMPTON RATIO AND EFFECTIVE ATOMIC NUMBER IN BIOLOGICAL SAMPLES

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ABSTRACT

The aim of this work was to develop a new method to correct the absorbed radiation (the mass attenuation coefficient curve) in low energy (E<30 keV) applied to a biological matrix based on the Rayleigh to Compton scattering ratio and the effective atomic number. For calibration, scattering measurements were performed on standard samples (H_3BO_3, Na_2CO_3, $CaCO_3$, Al_2O_3, K_2SO_4 and MgO) of radiation produced by a gamma-ray source of Am-241(59.54 keV) also applied to certified biological samples of milk powder, hay powder and bovine liver (NIST 1557B). In addition, six methods of effective atomic number determination were used as described in literature to determinate the Rayleigh to Compton scattering ratio (R/C) , in order to calculate the mass attenuation coefficient. The results obtained by the proposed method were compared with those obtained using the transmission method. The experimental results were in good agreement with transmission values suggesting that the method to correct radiation absorption presented in this paper is adequate for biological samples.

1. INTRODUCTION

The mass attenuation coefficient (μ/ρ) is an important parameter that describes the interaction of high-energy electromagnetic radiation with matter through photoelectric absorption, Compton scattering, Rayleigh scattering and pair production. For photon energies below 1

MeV the major interaction processes considered are incoherent (Compton) scattering, coherent (Rayleigh) scattering and photoelectric absorption.

The knowledge of mass attenuation coefficients of X-ray and gamma photons in biological material is of significant interest for industrial, biological, agriculture and medical applications such as dosimetry, radiography and computerized tomography [1, 2].

The mass attenuation coefficient provides a wide variety of information about fundamental properties of matter in the atomic and molecular level. It measures the probability of interaction of incident photons in matter per mass and area unit[2]. Several studies in literature seek accurate measurements of the mass attenuation coefficient of building materials [3, 4, 5], metal alloys, mineral samples [6, 7, 8, 9], and biological samples such as bone, muscle and fat [10, 11].

Experimental techniques to obtain the mass attenuation coefficient basically use the measure of the attenuation of the transmitted beam and the density of the sample. This method is called in this paper, transmission method, and is the direct application of Beer-Lambert equation. However, biological samples with small thickness attenuate the transmitted beam very little and the application of transmission techniques is not possible. A alternative for this experimental limitation could be the study of the radiation scattered by the sample to characterize the mass attenuation coefficient.

The measurement of the Rayleigh to Compton scattering ratio (R/C) between peaks produced by the sample is an alternative to the transmission method. Some works in literature use this technique for scattering tomography [14, 15] and the determination of effective atomic number [16, 17, 18, 19].

The parameter "effective atomic number" helps visualizing many physical characteristics of a material with a single number. The effective atomic number is a useful mean for the interpretation of the attenuation of X- ray or gamma radiation by a complex medium such as a biological tissue. Especially, studies on the interaction of low energy photons with biological samples are important for X-ray fluorescence and X-ray diffraction.
The method presented in this paper is based on the assumption that the radiation absorption by the sample can be represented by a power function of the energy E of the incident radiation on a biological matrix ($6 \leqslant Z \leqslant 15$ and $E < 30$ keV). This study proposes the development of a method for the determination of mass attenuation coefficients in samples with low atomic number based on Rayleigh to Compton scattering ratio and the effective atomic number.

2. THEORICAL CONSIDERATIONS

In interaction of photons with low-energy electrons very connected, the interaction can occur where the atom absorbs all the backtracking and practically no photon loses energy, simply changing its direction. This kind of interaction is called scattering Rayleigh or incoherent, and the direction of scattering is predominant forward [20]. The scattering of electrons bound to atoms is done fixing up the Thomson cross section for the electron free, considering the possibility of interference of scattered radiation. This correction appears as a Fourier transform of the density of charge, known as form factor [21].

$$\left(\frac{d\sigma}{d\Omega}\right)_R = \left(\frac{d\sigma}{d\Omega}\right)_{Th} .[F(x,Z)]^2 \quad (1)$$

where Z is the atomic number and x is the momentum transfer.
Unlike Rayleigh scattering, the Compton scattering, or incoherent, occurs from the interaction between a photon and a free electron. In this process the photon is completely absorbed. The result of this interaction is the emergence of another photon being on the direction of the original photon scattered in a direction. The photon transferred energy and momentum for the electron [22]. The Compton cross section is evaluated from:

$$\left(\frac{d\sigma}{d\Omega}\right)_C = \left(\frac{d\sigma}{d\Omega}\right)_{KN} .[S(x,Z)] \quad (2)$$

where Klein-Nishina refers to Compton cross section for a free electron at rest, while the incoherent scattering function *S(x,Z)* corrects for the fact that actually the electron is bound in an atom and moving.
The scattering Rayleigh and Compton are functions of the parameters of correction factor in *F(x,Z)* and the scattering function *S(x,Z)*, which depends on the momentum transfers and the atomic number. Thus, for given momentum transfers, the function S and F depend only of the atomic number. Obtaining the ratio of the cross section, we have:

$$\frac{\left(\frac{d\sigma}{d\Omega}\right)_R}{\left(\frac{d\sigma}{d\Omega}\right)_C} = \left\{\frac{\left(\frac{d\sigma}{d\Omega}\right)_{Th}}{\left(\frac{d\sigma}{d\Omega}\right)_{KN}}\right\} \times \frac{[F(x,Z)]^2}{[S(x,Z)]} \quad (3)$$

In the case of a solution containing n different elements, we can generalize the equation adding up to factor in *F(x,Z)* and incoherent scattering function *S(x,Z)* the percentage of each atomic element.

$$\frac{\left(\frac{d\sigma}{d\Omega}\right)_R}{\left(\frac{d\sigma}{d\Omega}\right)_C} = \left\{\frac{\left(\frac{d\sigma}{d\Omega}\right)_{Th}}{\left(\frac{d\sigma}{d\Omega}\right)_{KN}}\right\} \times \frac{\sum \alpha_i^{at} .[F(x,Z)]^2}{\sum \alpha_i^{at} .[S(x,Z)]} \quad (4)$$

Where:

$$\alpha_i^{at} = \frac{\left(w_i \big/ A_i\right)}{\sum\left(w_i \big/ A_i\right)} \quad (5)$$

α_i^{at} is the percentage of mass and w_i is the atomic number of each element A_i.

For a mixture, the ratio cross sections depends only effective atomic number Z_{eff}, which is a complicated function of atomic number present in each compound. Therefore, knowing it the

Rayleigh to Compton scattering ratio, we can get the curve of radiation absorption for a specific type of sample.

3. MATERIAL AND METHODS

3.1. Attenuation Coefficient

A beam of gamma radiation when focused on a material of thickness *D*, a fraction of the beam is absorbed by the material. The intensity of the beam that emerges is linked to the intensity I_0 of the incident beam, the Beer-Lambert law, being valid for a monoenergetic beam of radiation [22]:

$$I = I_0 . e^{-\mu D} \tag{6}$$

where μ is called the linear attenuation coefficient. The linear attenuation coefficient is the probability of suffering beam attenuation to the processes of photoelectric absorption, Compton scattering or pair production and can be written as:

$$\mu = \tau_{ph} + \sigma_C + \kappa_{pp} \tag{7}$$

The linear attenuation coefficient is limited by fact that it varies with the density of the absorber, even though the absorber material is the same. Therefore, the mass attenuation coefficient is much more widely used and is defined as:

$$\mu_m = \mu / \rho \tag{8}$$

where ρ represents the density of the medium. For a given gamma-ray energy, the mass attenuation coefficient does not change with the physic state of a given absorber.

$$I = I_0 . e^{-\mu_m . \rho . D} \tag{9}$$

For biological matrix and Z low energy ($6 \leqslant Z \leqslant 14$ and E < 30 keV) the μ_m can be represented by a function power of energy E of the radiation incident through the expression:

$$\mu_m = AE^B \tag{10}$$

where: E is in keV and A, B are constants.

Taking natural logarithms on either side of the equation 10, we have:

$$\ln(\mu_m) = \ln(A) + B\ln(E) \tag{11}$$

The equation 11 is linear function between the ln (μ) and ln(E). So, the constants A and B can be obtained by a simple linear regression analysis.

XCOM is the most popular program used to calculate photon cross sections for scattering, photoelectric absorption and pair production, as well as total attenuation coefficients, in any element, compound or mixture, at energies from 1 keV to 100 GeV [23]. The Figure 1 shows the feature of the mass attenuation coefficient curve to water in the range 1 to 100 keV using XCOM. We can observe the linear behavior in the range of 1 keV up to 30 keV.

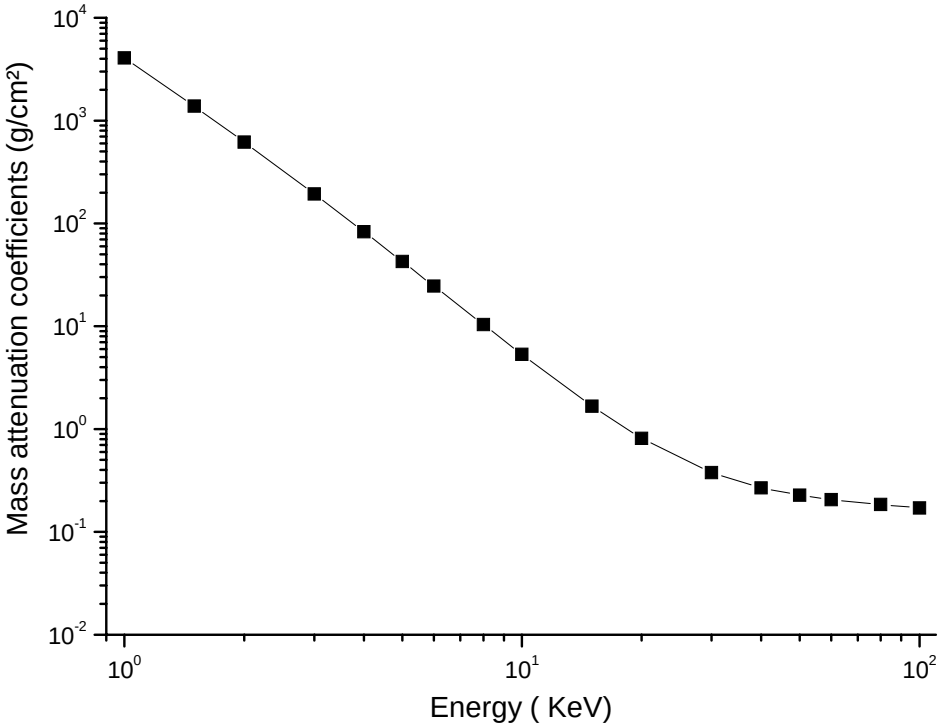

Figure 1. Mass attenuation coefficients curve to water in the range energy 1 to 100 keV.

The method of transmission used this work consist in determine experimentally of the mass attenuation coefficient through of the Beer-Lambert law. The method of transmission was applied for the energies of 13.95 keV, 17.74 keV, 22.12 keV and 26.36 keV.

3.2. The methods of determination effective atomic number

We can use reference samples with compositions very defined in order to we determine a curve that expresses the relation between R/C and Z_{eff} of the samples of references. So, starting from that curve we can determine Z_{eff} of an unknown sample. Recent works show that there is not a relation pre-defined to calculate the effective atomic number Z_{eff} of a composed material. Therefore, in the next step, we will be introduce some methodologies found in the literature for calculate the effective atomic number of a sample composed of several elements.

Six methods to determine the effective atomic number by Rayleigh to Compton scattering ratio in the literature were used in calculating the absorption coefficient. Below the description of each of the five methods:

3.2.1. Method I

In the first method, Harding [24] assumed that the ratio between Rayleigh to Compton scattering ratio is a function power of effective atomic number:

$$R = K.(Z_{eff})^{A} \tag{12}$$

where K represents the Thomson to Klein-Nishina cross section ratio and A is a power of $Zeff$. We can assume that the Rayleigh and Compton scattering are proportional to Z^3 and Z, respectively [25]. Thus, we get the following definition of Z_{eff}.

$$Z_{eff} = \left[\frac{\sum \left(w_i / A_i \right).Z_i^3}{\left(w_i / A_i \right).Z_i} \right]^{1/2} \tag{13}$$

3.2.2. Method II

The method propose for Duvauchelle *et al.* [26] use the atomic form factor $F(x,Z)$ and the incoherent scattering function $S(x,Z)$, as depends of the atomic number and of the momentum transfer x. For each value of the momentum transfers, there is a discrete function f_x that provides the value of Z as a function of F^2/S.

$$Z = f_x^D \left(F^2 / S \right) \tag{14}$$

For a sample formed by a composite of several elements, we can consider that the functions f_x are continuous and allow us calculate the value of Z_{eff}.

$$Z_{eff} = f_x \left[\frac{\sum \alpha_i^{at} .[F(x, Z_i)]^2}{\sum \alpha_i^{at} .S(x, Z_i)} \right] \quad (15)$$

$$Z_{eff} = f_x \left[\left(F^2 / S \right)_{eff} \right] \quad (16)$$

We can deduce the equation f_x through a set of appropriate curve. That way you can determine the value of Z_{eff} know the value of $(F^2/S)_{eff}$ of the sample considered.

3.2.3. Method III

Tsaï and Cho [27] suggest an empirical formula, very similar to the previous one, making use of the electronic percentage A. According to the authors, this expression is valid for energies below 150 keV.

$$Z_{eff} = \left(\sum_i \alpha_i^e .Z_i^{3.4} \right)^{1/3.4} \quad (17)$$

with

$$\alpha_i^e = \frac{\left(\frac{w_i}{A_i} \right) .Z_i}{\sum \left(w_i / A_i \right) .Z_i} \quad (18)$$

3.2.4. Method IV

According Puumalainen [28] the effective atomic number is the mean number of electrons per atom. Using the atomic percentage α_i^{at}, we can write equation below:

$$Z_{eff} = \sum_i \alpha_i^{at} .Z_i \quad (19)$$

3.2.5. Method V

This method, we obtained the effective atomic number using the empirical relation [29] given below:

$$Z_{eff} = \left[\sum_i f_i .(Z_i)^{2.94} \right]^{1/2.94} \quad (20)$$

where f_i is fraction of total number o electrons associated with each element and Z_i is atomic number of each element.

3.2.6. Method VI

Second the work of Manohara [30] the effective atomic number can be calculated for:

$$Z_{eff} = \frac{\sum f_i . A_i . (\mu_m)_i}{\sum f_i . \frac{A_i}{Z_i} . (\mu_m)_i} \tag{21}$$

Where $f_i = n_i \Big/ \sum n_i$ and Z_i are fractional abundance and atomic number of constituent element, respectively. n_i is the total number of atoms of the constituent element, $\sum n_i$ is the total number of atoms presents in the molecular formula.

4. EXPERIMENTAL PROCEDURE

The experimental measurements were conducted in two steps. In the first step, for calibration (R/C ratio versus Z_{eff}), the scattering measurements were performed on standard samples (H_3BO_3, Na_2CO_3, $CaCO_3$, Al_2O_3, K_2SO_4 and MgO) produced by a gamma-ray source of Am-241(59.54 keV). In addition, the same methodology was employed for all the certified biological samples (milk powder, hay powder and bovine liver) with Z_{eff} unknown. In the second step, transmission measurements were performed on reference samples using non-sealed Am-241 source. All samples were measured in triplicate.

4.1. Scattering set-up

The experimental set-up for scattering measurements was composed by: sealed Am-241 gamma source with an activity of 3.7 GBq emitting monoenergetic photons of 59.54 keV; an AMPTEK CdTe detector, with an energy resolution of about 0.53 keV at 14.4 keV and an ORTEC multichannel-analyzer. The angle between the emerging beam that reaches the detector and the surface of the sample was approximately 90° and the incident angle was 16°, creating a scattering angle of 106°. In the Am-241 source a lead collimator of 4.0 mm in inner diameter and 60 mm in length was used in order to narrow the incident beam. On the other hand, the detector was collimated with an aluminum collimator with 2.0 mm in diameter and 15 mm in length. Five different standard samples H_3BO_3, Na_2CO_3, $CaCO_3$, Al_2O_3, and K_2SO_4 were used to determine the calibration curve R/C vs. Z_{eff} . The reference sample MgO was not used in the calibration curve, but as reference sample to evaluate the scattering and transmission methods with the simulated results from XCOM. These samples were chosen because they have an atomic number between 5 and 16. Samples were prepared in pellets form with surface density of about 400 mg/cm² (25.4 mm diameter and thickness of 2.0 mm). Each measurement was performed during 5000 s. The reference samples were prepared in a similar way to the standard samples. Figure 2a shows the schematic view of the experimental scattering set-up.

4.2. Transmission set-up

The transmission measurements were applied only to the certified biological samples (milk powder, hay powder and NIST 1557B bovine liver). The experimental set-up used for transmission method was the same used for scattering measurements. However, the Am-241 gamma source used was non-sealed with an activity of approximately 370 MBq. In this configuration, the distance source-detector was 15 cm and an aluminum collimator with 1.0 mm in diameter and 15 mm in length was placed in front of the detector. The transmission method was applied for the energies of 13.95 keV, 17.74 keV, 22.12 keV and 26.36 keV. All the samples were prepared in pellet form with surface density of about 100 mg/cm². All transmission measurements were acquired in 300 s. The figure 2b shows the schematic view of the transmission experimental set-up.

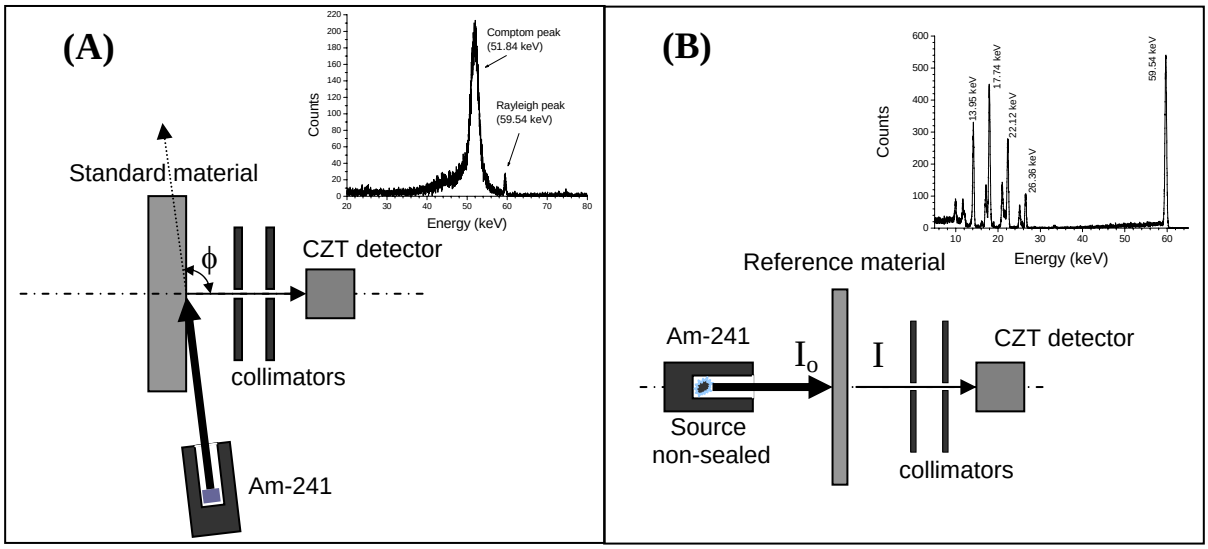

Figure 2. Scattering set-up (A) and Transmission set-up (B).

5. RESULTS AND DISCURSSION

Figure 2 shows de experimental setups and the energy spectra of Am-241 source (sealed and non-sealed source). It can be observed that the energy spectrum of scattering setup in the top of Figure 2a shows the Rayleigh and Compton scattering peaks for a low Z sample (H_3BO_3 – boric acid). It can be observed that in this spectrum the Rayleigh scattering peak is more intense than the Compton scattering peak. This happens because the cross section for Rayleigh scattering is proportional to Z^3 while the cross section for Compton scattering is closely proportional to Z. Thus, in a simple way, it can be considered that the Rayleigh scattering is predominant for heavy elements and the Compton scattering is important for light elements. In figure 2b (also on top) the energy spectrum of the non-sealed Am-241 source is shown, this is an optimum geometry for the CZT detector, showing a performance near 99% of efficiency for energy range. The energies of those photopeaks (13.95 keV, 17.74 keV, 22.12 keV and 26.36 keV) were chosen for transmission measurements.

Spectra were recorded by using the MAESTRO II, an MCA Emulation Software developed by ORTEC. Then, the X-ray spectrum was fitted and the areas under the photopeaks were evaluated (peak deconvolution and background subtraction) using the QXAS software package from IAEA[31]. The QXAS fitting procedures allow the spectral evaluation using either Gaussian or Voigt peak profiles.

The experimental results on the Rayleigh to Compton ratio (R/C) for standard and reference samples are shown in tables 1 and 2 respectively. In table 1 the values of Z_{eff} for each standard sample using the six methods presented in item 3.2 can be found. The variation coefficient (VC) for Z_{eff} varied from 2 % to 8 % for all samples except for H_3BO_3 (15 %). In addition, the uncertainties associated with R/C experimental results were lower than 20 %, in accordance with other results found in literature [32]. Thus, the relationship between R/C and Z_{eff} for each method studied for the standard samples using the data found in table 1 is graphically shown in Figure 3. The graphics in Figure 3 indicate that the best curve fitting was through linear regression ($R^2 > 0.98$) for Z_{eff} ranging from 5 to 16 for all the methods. Then, there is a relation between the R/C ratio and Z_{eff} that can be well described as a linear relation in the range of low Z_{eff}. Similar results were found by Leichter [16] and Donativi [32]. Besides that, the slope of each curve was practically constant over the limited range of effective atomic numbers which has been considered. The value of slop varied from 1.9×10^{-3} to 3.0×10^{-3} (fitted lines having a quasi identical slope).

Table 1. Results effective atomic number each method and Rayleigh to Compton ratio

Sample	Z_{eff}						Rayleigh to Compton ratio (R/C)
	Meth. I	Meth. II	Meth. III	Meth. IV	Meth. V	Meth. VI	
H_3BO_3	6.9	7.7	7.3	7.1	7.4	4.8	$2.3 \pm 0.6\ 10^{-3}$
Na_2CO_3	9.2	9.5	9.4	9.1	9.4	8.9	$6.1 \pm 0.8\ 10^{-3}$
Al_2O_3	10.8	11.1	11.2	10.6	11.1	10.5	$12.3 \pm 0.8\ 10^{-3}$
$CaCO_3$	13.9	13.0	15.5	12.6	15.1	13.4	$18.8 \pm 1.4\ 10^{-3}$
K_2SO_4	15.1	14.3	16.0	14.4	15.8	14.9	$21.7 \pm 1.5\ 10^{-3}$

The table 2 shows the Z_{eff} for three certified biological samples of unknown compositions (milk powder, hay powder and bovine liver) and one reference sample (MgO). The Z_{eff} values found in the table 2 were obtained from a linear relation between R/C and Z_{eff} for each method in Fig.3.

Table 2. Results experimental Rayleigh to Compton ratio and effective atomic number

Sample	Meth. I	Meth. II	Meth. III	Meth. IV	Meth. V	Meth. VI	R/C
MgO	10.8	10.9	11.4	10.4	11.3	10.2	$11.4 \pm 1.0\ 10^{-3}$
Bovine	7.2	8.0	7.4	7.4	7.5	5.5	$2.7 \pm 0.6\ 10^{-3}$
Milk	7.5	8.2	7.8	7.6	7.9	5.9	$3.5 \pm 0.8\ 10^{-3}$
Hay	8.4	8.9	8.7	8.4	8.8	7.0	$5.5 \pm 0.8\ 10^{-3}$

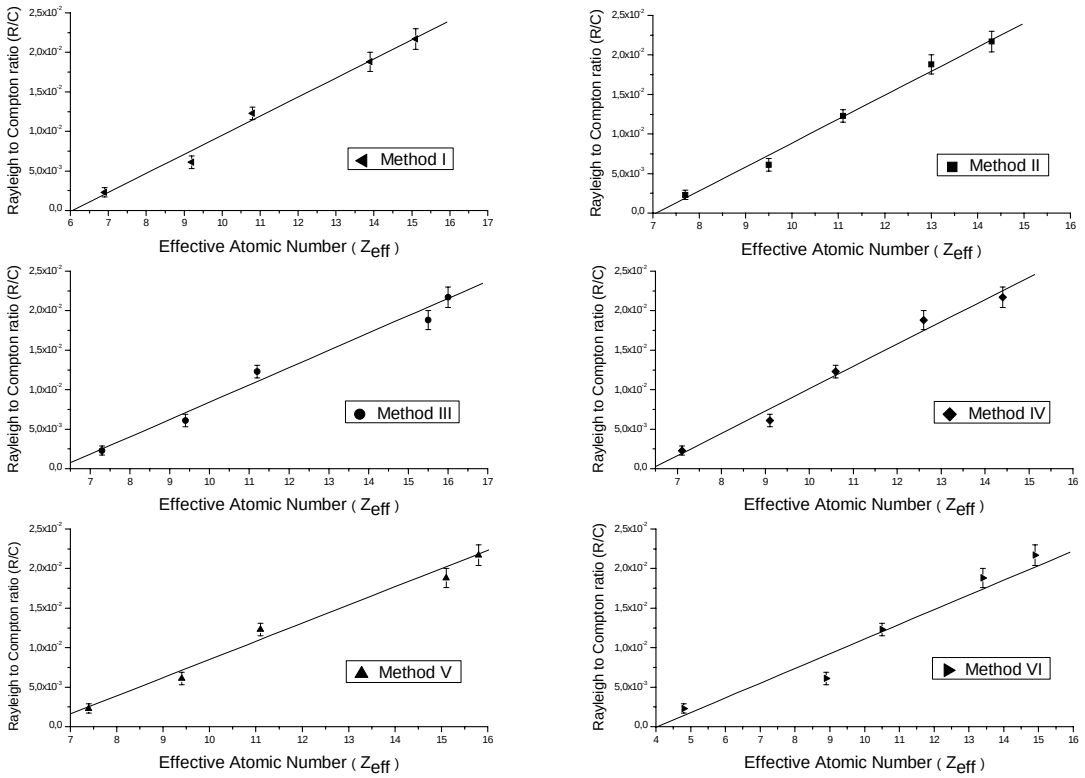

Figure 3. Relationship betweens peaks scattering Rayleigh to Compton ratio and effective atomic number for each method.

The validity of the method was tested comparing the values of Z_{eff} of different standard compounds determined using the experimental Rayleigh to Compton ratios. Plotting a curve of the Rayleigh to Compton ratio in each sample by effective atomic number, we can find the effective atomic number of a sample unknown element composition. The objective of these measures was to find the mass attenuation coefficient of the samples through the scattering produce by sample, and compare with the simulated values in XCOM.

The values found mass attenuation coefficients for six methods were the values founds for method of transmission. The figure 4 shows the results of the mass attenuation coefficients

found for each sample on the each energy of the source non-sealed. For sample MgO, figure 4d, the results were compared with results of the XCOM too.

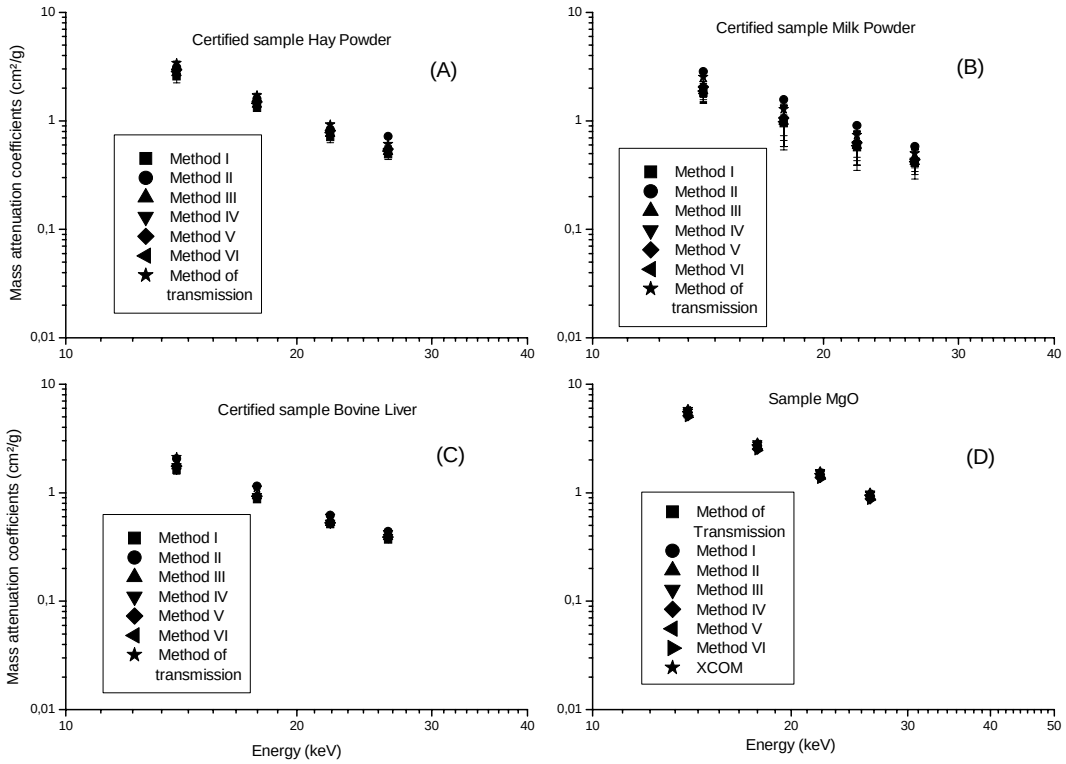

Figure 4. Results mass attenuation coefficients the samples Hay powder (A), Milk powder (B), Bovine powder (C) and MgO (D).

Comparing the values of the mass absorption coefficient obtained by the transmission method and the six methods for effective atomic number determination with the simulated results obtained by XCOM, demonstrate that the average percent error was below 5% for the sample MgO in the methods I (4.7%), II and III (1.2%), V (1.7%) and in the transmission method (5.0%). Methods IV (9.6%) and VI (16%) did not present satisfactory results. For certified samples Hay powder, Milk powder and Bovine liver, it was not possible to simulate the values of the mass absorption coefficient by XCOM, because the compositions of these materials are not known. Thus, the values found by the method proposed in this paper were compared with the values obtained by the transmission method. In this comparison, method II showed the best results with average percent error of 2.7% for the Hay powder sample, 8.8% for the Bovine liver sample and 16% for the milk powder sample.

6. RESULTS AND DISCUSSION

The proposed methodology efficiently showed that attenuation coefficient results can be obtained in a critical range of energies (2-30 keV) with uncertainties below 5% when compared with the method of radiation transmission and with XCOM. The application of the method is fast and easy, being only necessary the use of a calibration curve which is obtained from a small number of standards and the determination of the effective atomic number of the analyzed sample using the ratio R/C. Thus, the proposed methodology can be a useful tool, mainly in situations where the composition of the sample is unknown and the transmission method unfeasible as in the cases of analytical techniques that use X-ray spectrometry. The method for Z_{eff} determination that presented the best final result in the energy of incident beam used in this study (59.54 keV) was Method II

The next stage of our study will include the application of the methodology to other energy ranges using tubes with anodes of Mo and Rh and in different scattering angles to assess the limitations of the technique and whether method II still presents the best results.

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