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Mass attenuation coefficients of X-rays in different barite concrete used in radiation protection as shielding against ionizing radiation

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ABSTRACT

The probability of a photon interacting in a particular way with a given material, per unit path length, is usually called the linear attenuation coefficient (μ), and it is of great importance in radiation shielding. Plates of barite concrete with different thickness were fabricated in order to determining their mass attenuation coefficients at different energies. The plates were irradiated with ISO X-ray beams (N60, N80, N110 and N150), generated by Pantak HF320 X-ray equipment, at the IPEN laboratory. The mass attenuation coefficients of barite concrete have been measured using X-ray attenuation for different thicknesses of barite concrete qualities of the ISO. The attenuator material issued from different regions of Brazil. The experimental procedure in this research was validated by comparison between the experimental measurements of mass attenuation coefficients and coefficients determined by the same atomic composition, using as a tool to XCOM. The highest value of (μ/ρ) found experimentally was in the energy of 48 keV, in ISO 60 N quality, being $1.32(\pm 0.49)$ for purple barite; $1.47(\pm 0.41)$ for white barite and $1.75(\pm 0.41)$ for cream barite. The determination of the chemical composition of the barite samples was of fundamental importance for the characterization of these materials. It can be seen that both calculated and measured data for the linear attenuation coefficients increase with the increasing materials density, as it is expected. It can be concluded that the photon attenuation coefficients depends on the photon energy and the materials density is the main contribution to the photon attenuation coefficients, which is important for radiation shielding.

1. Introduction

Concrete which contains water, cement and aggregate, is widely used in building constructions such as medical hospitals. The acknowledgement of the characteristics of attenuation of barite is fundamental, from the radioprotection point of view to the viabilisation, the design and the execution of projects of ionizing radiation shielding in radioactive facilities. Planning the appropriate shielding for nuclear centers depends on several factors such as the type of radiation, energy and cost. Mostly high atomic number and high density materials are used for this purpose; but the latter are costly, hence their use is limited. The use of ionizing radiation in medical and dental diagnosis is supervised by the sanitation department in both municipal and state levels, in accordance to rules established by the National Agency of Sanitary Supervision in Brazil - ANVISA. Moreover, The Ministry of Labour and Employment also inspects the workplaces, with the aim to control occupational exposures. For each composite materials of several interests, for X-ray and gamma– photon interactions, is assigned a number (equivalent to atomic number in elements) known as “effective

atomic number”, as well as X-ray mass attenuation coefficients. [Singh et al. (2010); Nicholas (2010); Morabad and Kerur 2010; MS, 1998).

The walls and doors, in an X-ray equipment room, must have their thickness accurately calculated as to assure protection of the public in general as well as the staff occupationally exposed to ionizing radiation. In practice, the dimensioning of the barite mortar and concrete used in building and plastering walls is determined by the principle of thickness equivalence in relation to concrete or lead. The shielding properties of this material for X-rays are presented in terms of the linear attenuation coefficient μ (cm^{-1}) and it is defined as the probability of a beam interacting with a material per unit path length. An accurate measurement of the linear attenuation coefficient was developed by Kerur et al. (1991). For the authors the best results were obtained for those thicknesses which satisfy $2 \leq \ln(I_0/I) \leq 4$, corresponding to transmission (T) range of $0.5 \geq T \geq 0.25$. Since I_0 the incident X-ray beam and I the transmitted X-ray beam. A plot of the logarithm of transmission as a function of specimen thickness yielded a straight line in each case, indicating the validity of Beer-Lambert's law [Kerur et al. (1991)].

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X-ray mass attenuation coefficients have a wide range of applications in X-ray science. Many theoretical calculations have been made. However, significant discrepancies exist between theory and theory, theory and experiment and experiment and experiment. Large discrepancies have been seen between the measurements and the theories of XCOM (Berger, 1999), in the region between the K-edge and up to about 2.5 keV above the edge. [Nicholas (2010)].

Several authors have studied (Shirmardi et al. (2013); Stankovic et al. (2010); Polat and Orhan, 2010) shielding properties of various barites concretes for gamma energies 0.662, 1.173, and 1.332 MeV using the MC code and compared to predictions from the XCOM code and experimental data.

Our study was conducted to evaluate the efficacy of different mixtures of barite concrete for shielding in diagnostic X-ray rooms.

2. Material and methods

This study aims at characterizing samples of purple, white and cream barite occurring in different parts of Brazil, which are used in radioprotection such as X-ray shielding materials. The analyses were performed at the laboratories of the Center of Development of Nuclear Technology (CDTN/CNEN). Their chemical compositions were determined by X-ray Fluorescence (EDX). The experimental procedure employed in this research to determine the mass attenuation coefficient (μ/ρ) was the use of ISO N (ISO, 1996) X-ray qualities, (N60, N80, N110 and N150), generated by Pantak HF320 X-ray equipment, at the IPEN/CNEN laboratory (Table 1). The beam of photons transmitted went through another collimator and reached an X-ray detector - type CdTe. This detector was calibrated for sources of ^{241}Am , ^{133}Ba , ^{109}Ca , for photon energies emitted according to Table 2. The measurements of attenuation of the X-ray beams were obtained in good geometry conditions, with a narrow beam, aiming at reducing the quantity of scattered radiation reaching the detector (Almeida et al., 2016; Almeida. et al. 2015). Tungsten collimators ranging from 25 to 2000 μm in diameter and thickness between 1 and 2 mm were used. The collimators were used by the detector, in order to reduce the radiation intensity at the detector, avoiding stacking pulses.

The measurements of the spectra were checked with the detector placed at 340 cm from the focal point, and the barite mortar plates or a set of them were positioned at 50 cm from the focal point. Reference X-ray qualities were used as recommended by ISO (ISO, 1996). By integrating the incident spectrum and the transmitted spectrum over selected width of the photo peak, incident intensity I_0 and transmitted intensity I were obtained. Finally the μ/ρ was obtained from the slope of the straight line fitted by plotting a graph of $\ln I$ as a function of thickness; method of least squares, following equation:

$$I = I_0 e^{-\mu/\rho(t)} \quad (1)$$

The I_0 denotes the photons intensity with energy, without attenuation and I is the photons with energy after attenuation; μ/ρ (cm^2/g) is the mass attenuation coefficient and t (g/cm^2) is the sample mass thickness (the mass per unit area).

The experimental procedure in this research was validated by the

Table 1
ISO X-ray reference radiation (Narrow spectrum).

Radiation quality	Total filtration	Effective energy (keV)	HVL (mm Cu)
N - 60	4 mmAl+0.6 mm Cu	48	0.25
N - 80	4 mmAl+2 mm Cu	65	0.61
N - 100	4 mmAl+5 mm Cu	83	1.14
N - 150	4 mmAl+2.5 mm Sn	118	2.40

Source: used in the calibration of the X-ray detector.

Isotope	Photon energy (keV)	Count Channel Number
^{241}Am	13.9	68.6
	17.8	83.4
	59.5	293.7
^{133}Ba	30.8	152.1
	80.9	399.9
^{109}Ca	22.1	108.8
	88.0	434.9

comparison between the experimental measurements of mass attenuation coefficients and coefficients determined by the same atomic composition, using XCOM (Berger et al., 1998; Saloman et al., 1988) and FFAST (Chantler, 2000, 1995).

(μ/ρ) is mass attenuation coefficient for materials composed of various elements; one may assume that the contribution of each element to the total interaction of the photon is additive “Mixture Rule”. In accordance to this rule, the total mass attenuation coefficient of a compound is the sum of the weight proportion of each individual atom present in it (Morabad and Kerur, 2010). Therefore:

$$\left(\frac{\mu}{\rho}\right)_{comp} = \sum w_i \left(\frac{\mu}{\rho}\right)_i \quad (2)$$

In which: (μ/ρ)_{comp} is the mass attenuation coefficient for the compound, (μ/ρ)_i is the mass attenuation coefficient for each individual element and w_i is the fractionated weight of the elements in the compound.

The procedure adopted for the determination of the mass attenuation coefficient (μ/ρ) was described by Kerur et al. (2009); Morabad and Kerur (2010); Zenóbio et al. (2011) and Teerthe and Kerur (2016). The range of transmission chosen for linear regression and the determination of μ/ρ should be such that Beer–Lambert’s law is rigorously valid. For a solid state detector, Creagh and Hubbel (1987) suggested the transmission range to be $0.02 < T < 0.13$, for proportional counter. Kerur et al. (1991) recommended the transmission range to be $0.25 < T < 0.5$ and for the NaI(Tl) detector to be $0.02 < T < 0.5$. So it would be of interest to find out the range of transmission for the CdTe detector in which Beer–Lambert’s law is rigorously valid. We plot the logarithm of transmission as a function of thickness for the range of transmission between 8% and 70% (Fig. 2, in results). With these standardized experimental parameters we, then determine μ/ρ for the remaining materials. The solid line in each figure is the least squares straight line fit.

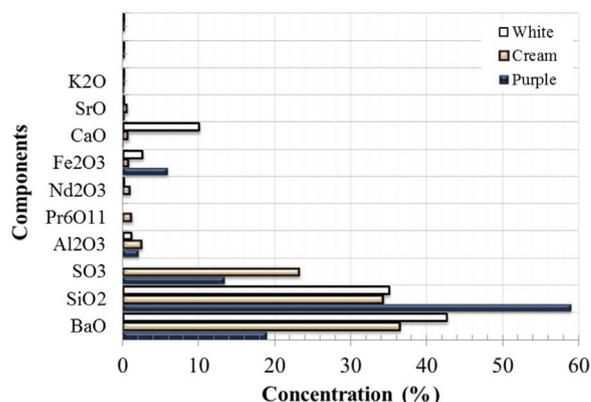


Fig. 1. Chemical elements found in the samples of this study. Analyzed by X-ray fluorescence.

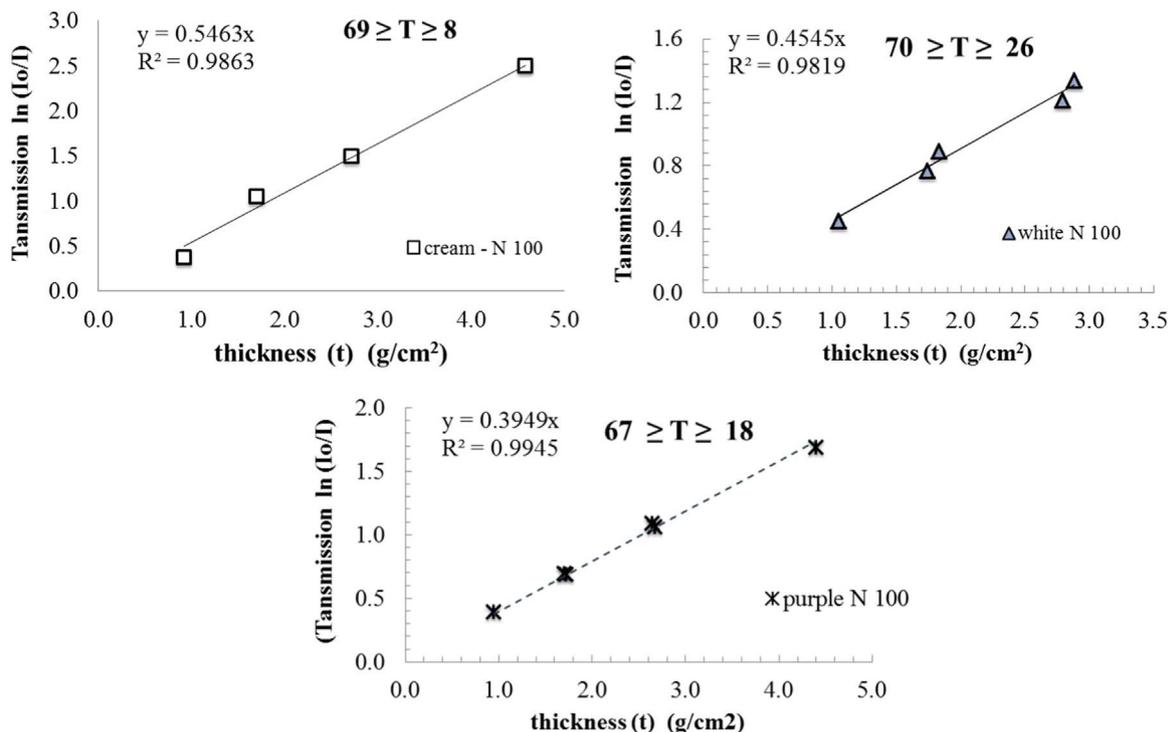


Fig. 2. Graph $\ln(I_0/I)$ versus thickness for transmission barites white, cream and purple. Irradiated with ISO X-ray beams, energy from 83 keV and with attenuation of 8–70%.

The maximum errors in the mass attenuation coefficients were calculated from errors in incident (I_0) and transmitted (I) intensities and areal density (t) as well as statistical counting (Ozdemir et al., 2009; Zenóbio et al., 2016).

$$\Delta\left(\frac{\mu}{\rho}\right) = \frac{1}{t} \sqrt{\left(\frac{\Delta I_0}{I_0}\right)^2 + \left(\frac{\Delta I}{I}\right)^2 + \left(\ln\frac{I_0}{I}\right)^2 + \left(\frac{\Delta t}{t}\right)^2} \quad (3)$$

3. Results and discussion

Plates of barite concrete having different thicknesses were fabricated in order to determine their mass attenuation coefficients at different energies. The attenuator material is used from different regions of Brazil, as follows: white barite mortar from the State of Paraíba, Brazil; cream barite mortar from the State of São Paulo, Brazil and purple barite mortar from the State of Bahia, Brazil. Observing that, from the materials studied in this research, the barite mortar of the State of Bahia presents the greatest difficulty in the extraction process. The determination of the chemical composition of the barite samples was of fundamental importance for the characterization of these materials. As a very reproducible, fast and accurate instrumental technique, X-ray fluorescence (EDX) is the most usual method for determining the chemical composition of materials (Barba et al., 1997). Among the barite samples studied, white barite, because it presents a higher percentage of barium, is the best raw material for the study of the feasibility of the manufacture of protective barriers, with shielding properties against X radiation. For a better comparison and visualization, these results are presented in Fig. 1.

From the fraction of the intensity transmitted, the incident air kerma was calculated for each thickness of the materials. The experimental (μ/ρ) values for the three materials were calculated using the measured values of I_0 and I and Beer-Lambert's law ($\mu/\rho = \ln T/t$). The respective inclination of the line is (μ/ρ) , which expresses the neperian logarithm (\ln) of the intensity transmitted against the thickness of the barite mortar. The attenuation coefficient μ/ρ was then calculated using the least squares fit method (Fig. 2) and the results are given in Tables 3–5.

Table 3

Experimental values of μ/ρ in cm^2/g obtained at different transmission (T) regions using least squares fit methods. White barite ($\rho = 3.10 \text{ g/cm}^3$).

Transmission, T (%)	Effective energy (keV)	(μ/ρ) Expt (cm^2/g)	(μ/ρ) theory (cm^2/g) XCOM	Dif. (Expt-theory)%
33 to 02	48	1.47 (± 0.41)	1.42	03.50
55 to 08	65	0.75 (± 0.27)	0.67	11.94
72 to 08	83	0.45 (± 0.14)	0.36	25.00
59 to 27	118	0.23 (± 0.14)	0.18	27.78

Table 4

Experimental values of μ/ρ in cm^2/g obtained at different transmission (T) regions using least squares fit methods. Purple barite ($\rho = 2.96 \text{ g/cm}^3$).

Transmission, T (%)	Effective energy (keV)	(μ/ρ) Expt (cm^2/g)	(μ/ρ) theory (cm^2/g) XCOM	Dif. (Expt-theory)%
26 to 3	48	1.32 (± 0.49)	0.99	32.32
49 to 6	65	0.67 (± 0.14)	0.48	39.58
67 to 18	83	0.39 (± 0.13)	0.27	44.44
70 a 29	118	0.20 (± 0.06)	0.14	42.85

Table 5

Experimental values of μ/ρ in cm^2/g obtained at different transmission (T) regions using least squares fit methods. Cream barite ($\rho = 2.99 \text{ g/cm}^3$).

Transmission, T (%)	Effective energy (keV)	(μ/ρ) Expt (cm^2/g)	(μ/ρ) theory (cm^2/g) XCOM	Dif. (Expt-theory)%
33 to 01	48	1.75 (± 0.41)	1.61	08.60
51 to 02	65	0.87 (± 0.23)	0.75	16.00
69 to 08	83	0.55 (± 0.15)	0.41	34.14
83 to 27	118	0.28 (± 0.10)	0.20	40.00

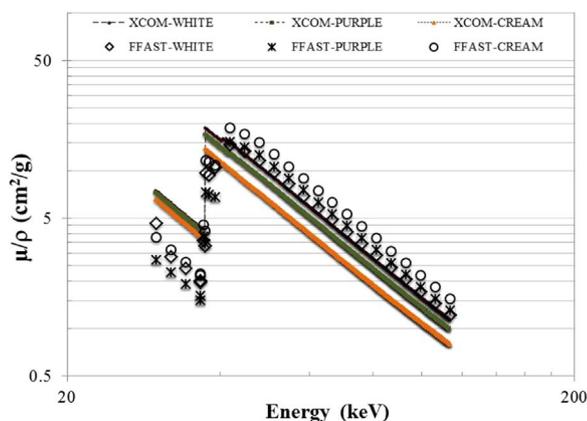


Fig. 3. Distribution of mass attenuation coefficient as a function of effective energy for mortar barite by Win XCOM programmer (Berger et al. (1998) and FFAST (Chantler, 2000, 1995).

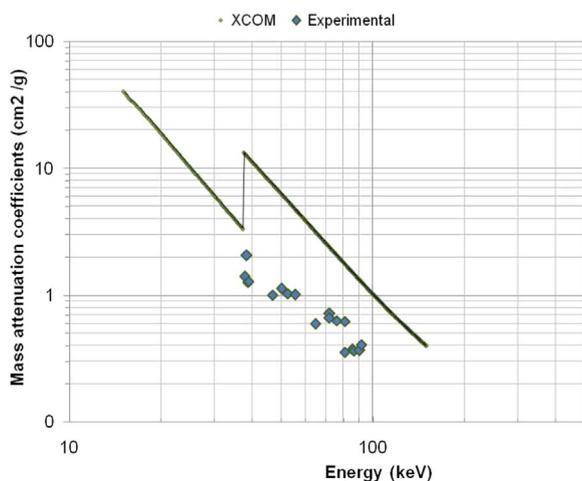


Fig. 4. Distribution of mass attenuation coefficient for photon energies for barite purple, experimental and theoretical by XCOM (Berger, 1999).

The experimental procedure in this research was validated by the comparison between the experimental measurements of mass attenuation coefficients and coefficients determined by the same atomic composition, using XCOM (Berger et al., 1998; Saloman et al., 1988) and FFAST (Chantler, 2000, 1995). The greatest discrepancies between these theories occur near the edges, with deviations by factors of 2 or more between the results. The cause of near-edge error in theoretical computations is often due to inadequate interpolation, extrapolation or integration methods, which introduce apparent oscillations or discontinuities into the data (Chantler, 1999).

The highest value of (μ/ρ) found experimentally was in the energy of 48 keV (see Tables 3–5), being $1.32(\pm 0.49)$ for purple barite; $1.47(\pm 0.41)$ for white barite and $1.75(\pm 0.41)$ for cream barite. In Figs. 3 and 4 the jump of (μ/ρ) corresponds to the absorption edge K of high atomic number elements which is defined as the minimum photon energy or the maximum wavelength that an electron can be expelled from a given energy level of the atom. The photon interacts with each element of the sample individually and the (μ/ρ) expresses the information of the sample as a whole. The graph of $\ln(E) \times \ln(\mu/\rho)$ is a straight line, showing the variation of (μ/ρ) with energy (Figs. 3 and 4). For this linear variation it is necessary that in the chemical composition of the sample there are no elements present whose absorption K is close to the energy of the incident electron (Morabad and Kerur, 2010; Polad, 2010). In our research it could be observed

that near the absorption edges of high atomic number elements the radiation attenuation properties of the investigated materials varied due to the presence of Ba, for example, as constituents of the sample studied (Fig. 1). Barium is the most common and abundant chemical element present in the compound with a percentage of around 62% for the white barite.

This study shows that the differences between the values of theoretical results and experimental ones, for mass attenuation coefficients, may be attributed to variation in the chemical composition (Fig. 1) of the samples and to the nature of the rule of mixtures, which disregards interactions amongst the atoms of the compounds. The Win XCOM program is based on the rule of mixture which shows the coefficients of attenuation of any substance, such as the sum of pondered contributions of the individual atoms (Berger, 1998). The measured X-ray mass attenuation coefficients of this work are compared to the FAST and XCOM calculated values and found with discrepancies increasing towards lower energies, as shown in Figs. 3, 4 and 5. This is not a linear graph because it indicates that the sample contains high atomic number elements. The highest atomic number elements present in the sample have a bond at layer K, close to the incident photon binding energy, in which case the mass attenuation coefficients of the X-rays are reflected in the graph, showing a linearity deviation, mainly in the vicinity of 37 keV energy.

The value calculated at each thickness varies with the thickness of the specimen at all thicknesses except at 50–25% transmission region (Kerur, et. al, 1999; Zenobio et al. 2016). The Beer-Lambert's law is rigorously valid in this region and the measured values agree closely with the theoretical value. In Table 6 the experimental μ/ρ values for cream barite, calculated using the measured values of I_0 , and I and Beer-Lambert's law, $(\mu/\rho = -\ln T/t)$ for different specimen thicknesses, are given in column 4. Fig. 2 show that at each transmission region, the experimental points lie close to the least squares fitted line, indicating the suitability of our method for measuring μ/ρ values. The least squares fit value of μ/ρ in calculated at each transmission. In Table 6, it is clear that our method for determining μ/ρ yields a 'best value' in the transmission region 50–25% which agrees closely with the theoretical value, as it can be seen in Tables 3–5.

The maximum relative expanded uncertainty ($k = 2.03$) of the experimental results for attenuation coefficients for barite: white, cream, purple was 15%, 12% and 8% respectively, to a confidence level of 95%.

4. Conclusion

It was possible to show that the characteristics of attenuation of barite influence the transmission curves within reach of the quality X-rays. Barium is the most common and abundant chemical element present in the compound with a percentage of around 62% for the white barite. It was also demonstrated that the composition of the barite influences in the mass attenuation coefficients of the X-rays, thus demonstrating the importance of the determination of the atomic composition of these materials. The determination of these quantities allows for higher reliability, precision and safety in the use and employment of the materials studied. The measured X-ray mass attenuation coefficients of this work are compared to the FAST and XCOM calculated. The greatest discrepancies between these theories occur near the edges, with deviations by factors of 2 or more between the results. It was also demonstrated that our method for determining μ/ρ yields a 'best value', in the transmission region 50–25%, agrees closely with the theoretical value.

It can be concluded in this work that the differences between the values of theoretical and experimental results for the mass attenuation coefficients can be attributed to the variations in the chemical composition of the samples and the nature of the mixing rule that neglects interactions between the atoms of the analyzed compounds.

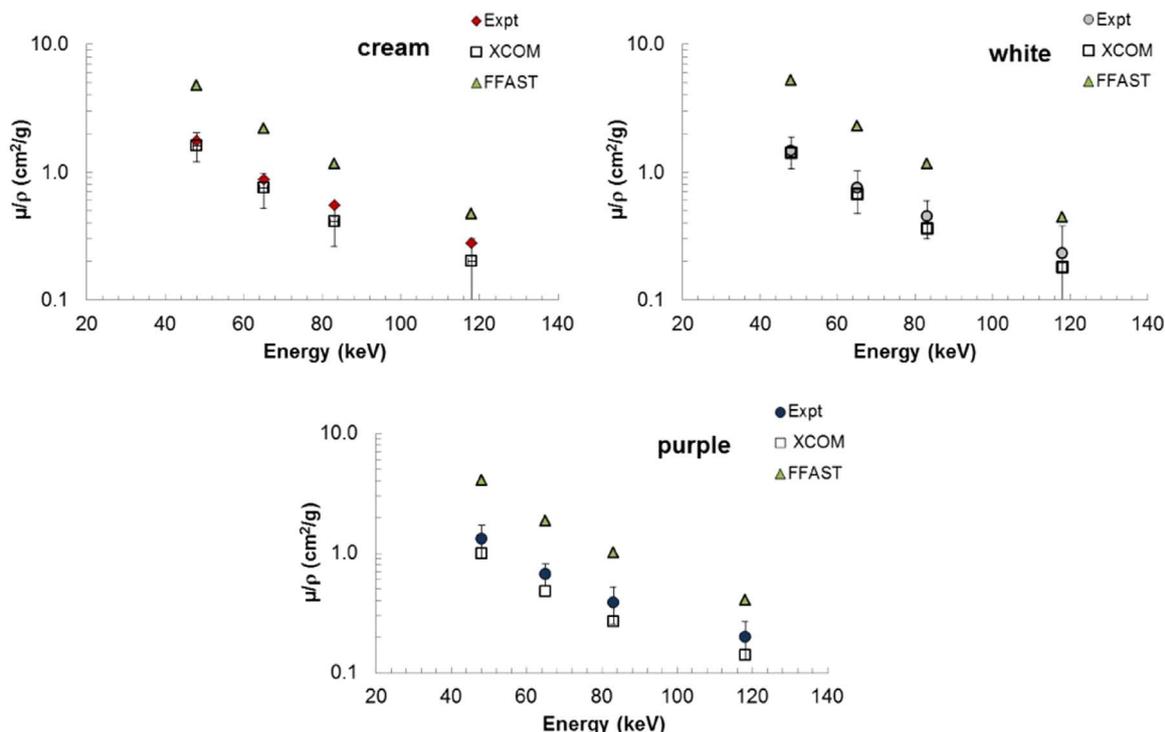


Fig. 5. Attenuation coefficients μ/ρ in cm^2/g , obtained for four qualities ISO for barites cream purple and white.

Table 6

Experimental values of μ/ρ in cm^2/g determined for thicknesses of Barite cream absorber in the energy of 83 keV (quality ISO-N100).

Thickness (t) (g/cm ²)	Kerma (mGy)	Transmission (T = I _o /I)%	$\mu/\rho =$ (ln T/t) (cm ² /g)- Expt.	μ/ρ (cm ² /g)- theory	Dif. (Exp.- theory)%
0.851	3.96	72.55	0.377	0.405	-2.737
0.885	2.87	71.35	0.381	0.405	-2.325
0.923	2.83	69.24	0.398	0.405	-0.642
1.873	2.74	30.79	0.629	0.405	22.415
1.797	1.22	32.38	0.627	0.405	22.276
2.558	1.28	26.09	0.525	0.405	12.066
2.724	1.03	22.48	0.548	0.405	14.321
4.593	0.89	8.28	0.542	0.405	13.762

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