

Gamma Ray Attenuation Coefficients for Lead Oxide and Iron Oxide Reinforced In Silicate Glasses as Radiation Shielding Windows

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Received in :1February 2014 Accepted in :2June 2014

Abstract

In this work, the mass attenuation coefficient, effective atomic number and half value layer parameters were calculated for silicate (SiO_2) mixed with various levels of lead oxide and iron oxide as reinforced materials. SiO_2 was used with different concentrations of PbO and Fe_2O_3 (25, 50 and 75 weight %). The glass system was prepared by the melt-quenching method. The attenuation parameters were calculated at photon energies varying from 1keV to 100MeV using the XCOM program (version 3.1). In addition, the mass attenuation coefficient and half value layer parameters for selected glass samples were experimentally determined at photon energies 0.662 and 1.28 MeV emitted from radioactive sources ^{137}Cs and ^{22}Na respectively in a collimated narrow beam geometry set-up using 2"x2" NaI (Tl) scintillation detector. These values are found to be in agreement with the values computed theoretically. Moreover, these results were also compared with those for the commercial window glass. The effective atomic number (Z_{eff}) and half value layer (HVL) results indicate that $\text{PbO}+\text{SiO}_2$ was better gamma ray attenuation than $\text{Fe}_2\text{O}_3+\text{SiO}_2$ and commercial window glass. This indicates that $\text{PbO}+\text{SiO}_2$ glasses can be used as gamma ray shielding in replace of both of them in this energy range.

Keywords: Gamma ray, mass attenuation coefficient, linear attenuation coefficient, effective atomic number, half value layer, glass, radiation shielding materials.

Introduction

Gamma-ray and X-ray attenuation coefficients are very important in both fundamental and applied science. They are invaluable in many applied fields, such as nuclear diagnostics, radiation protection, nuclear medicine, and radiation dosimetry. Protection of the body from unnecessary radiation exposure when working in a radiation area is a priority for every health physicist [1, 2]. Glass materials are possible alternatives for radiation shielding materials with two advantages brought by their transparency to visible light, and their properties can be modified by using composition and preparation techniques. Silicate glasses are the most commonly available commercial glasses due to ease of fabrication and excellent transmission of visible light [1,2]. Lead oxide (PbO) is a promising gamma ray shielding materials due to its strong absorption of gamma rays [3, 4]. Authors has explored the possibility of using glass as gamma ray shielding material in terms of heavy metal-silicate glasses in their research article[5, 6]. In photon interaction with composite materials, the atomic number cannot be represented uniquely by a single number across the entire energy region, as in the case of pure elements. This number for composite materials is called the effective atomic number and it varies with the photon energy [7]. Berger and Hubbell [8] have developed a computer program (XCOM) which calculates photon cross-sections and attenuation coefficients for pure elements and mixtures in the energy range of 1 keV to 100 GeV. In the present study, the shielding parameters such as mass attenuation coefficient (μ_m), effective atomic number (Z_{eff}) and half value layer (HVL) were calculated for lead-silicate (PbO -SiO₂) and iron-silicate (Fe₂O₃-SiO₂) glasses containing the same levels of concentration at photon energies varying from 1keV to 100MeV by using the (XCOM) program . Also, we measured the mass attenuation coefficient and half value layer of gamma-rays for PbO-SiO₂ and Fe₂O₃-SiO₂ glasses at 0.662 and 1.28 MeV photons by using NaI(Tl) scintillation detector. Lead silicate and iron silicate glasses were synthesized by using a melt quenching method.

Theory

The theoretical relations used in the present work are summarized in this part. A collimated beam of mono-energetic gamma ray is attenuated in matters according to the Lambert-Beer law [9,10]:

$$I = I_0 e^{-\mu x} \quad (1)$$

Where I_0 is the initial intensity of gamma ray, I is the intensity of gamma ray after attenuation through a material of thickness x (cm) and μ is the linear attenuation coefficient (cm⁻¹) of the material. Mass attenuation coefficient (μ_m) of the material is obtained by dividing μ by the material density (ρ). The mass attenuation coefficient, for a compound or mixture is given by [9,10]:

$$\mu_m = \sum_i w_i (\mu_m)_i \quad (2)$$

Where w_i and $(\mu_m)_i$ are the weight fraction and mass attenuation coefficient of the i th constituent element, respectively. For any compound, the total atomic cross section (σ_a) can be calculated from the knowledge of mass attenuation coefficient by the following formula [7, 11,12]:



$$\sigma_a = \frac{\mu_m}{N_A \sum_i \frac{w_i}{A_i}} \quad (3)$$

Where N_A is Avogadro's number and A_i is the atomic weight of i th element. Similarly, the total electronic cross section (σ_{el}) is given by [7, 11, 12]:

$$\sigma_{el} = \frac{1}{N_A} \sum f_i \frac{A_i}{Z_i} (\mu_m)_i \quad (4)$$

Where f_i denotes the fractional abundance of the element i with respect to the number of atoms such that $f_1 + f_2 + f_3 + \dots + f_i = 1$, Z_i is the atomic number of i th element. Finally, by using Eqs. (3) and (4), the effective atomic number (Z_{eff}) can be defined as:[7,11,12]:

$$Z_{eff} = \frac{\sigma_a}{\sigma_{el}} \quad (5)$$

The thickness of the material that reduces the photon beam intensity to half of its original value (I_0), i.e. $(\frac{1}{2}) I_0$, is called the half value layer (HVL) and is given by [9]:

$$HVL = \frac{\ln 2}{\mu} = \frac{0.693}{\mu} \quad (6)$$

where μ is linear attenuation coefficient of the material at a given photon energy.

Materials and methods

Sample preparation and density measurements

Starting materials of (99.9% purity) were used in the present work for Fe_2O_3 , PbO and SiO_2 . All the chemicals were weighed accurately using an electrical balance with an accuracy of 0.001g, grounded to fine powder and mixed thoroughly. The samples were prepared by mixing of PbO and Fe_2O_3 in concentrations of 25, 50 and 75 (weight %) with SiO_2 . Each batch of about 50 g in alumina crucible was melt in an electrical furnace for one hour, at $1250^\circ C$. The melts were then poured between the stainless steel molds. The quenched glasses were annealed at $500^\circ C$ for 3 hours to reduce thermal stress, and cooled down to the room temperature. At the room temperature, densities of glass samples were measured with the Archimedes' method using xylene as an immersion liquid. The density of glass sample (ρ) was calculated by the formula:

$$\rho = \frac{W_A}{W_A - W_B} \times \rho_L \quad (7)$$

Where W_A and W_B are the weight of the sample in air and the weight of the sample in xylene, respectively and ρ_L is density of xylene. The chemical compositions, % by weight, and the density of the glass samples are given in Table 1. Fig.1 shows density plots of the glass for both systems. It is seen that the density of the glass samples increases with higher Fe_2O_3 and PbO contents, because of higher molecular weight of Fe_2O_3 and PbO in comparison to SiO_2 . The $PbO+SiO_2$ glass samples prepared in our work gave higher densities than the $Fe_2O_3+SiO_2$ glasses over all ranges of SiO_2 concentration which may be contributed to higher atomic weight of the lead.

Calculation of the total mass attenuation coefficients

The theoretical values of mass attenuation coefficient (μ_m) were calculated using the XCOM computer program (version 3.1). The used XCOM program have been recently

modified to calculate the total mass attenuation coefficients for elements, compounds and mixtures at photon energies varying from 1 keV to 100 GeV [8], and provides total cross section as well as partial cross sections for various interaction processes.

Gamma ray measurements by NaI (Tl) scintillation detector

Experimental measurements have been performed to investigate attenuation values of gamma rays for the six Fe₂O₃, PbO / SiO₂ glass samples. The present experimental arrangement can be identified as being of narrow beam attenuation geometry which gives gamma ray buildup factor equal to unity. The gamma ray spectrometer is energy calibrated using standard multi energy gamma sources : mix source ¹³⁷Cs (has photo peak 662keV) with ²⁴¹Am (has photo peak 59.5keV) manufactured by LD.G.mbh-Germany. in addition using ⁶⁰Co (have photo peaks 1.17 and 1.33 MeV). The diagram of experimental setup is shown in Fig. 2a. All samples were used in the form of a tablets plate with 2 cm diameter and thickness 0.4 cm. Samples with different thicknesses (0.4 -1.6) cm were arranged in front of a collimated beam emerged from radioactive source. Two standard radioactive gamma sources ¹³⁷Cs (0.662 MeV) of 71.62μCi (2.65 MBq) strength and ²²Na (1.280 MeV) of 0.445 μCi (16.47 kBq) strength were used. The intensities of gamma photons were measured by using 2"x2" NaI (Tl) scintillation detector (Saint- Gobain Crystals Bicron). The detector was coupled to pre-amplifier, amplifier, power supply and computer analyzer with LD Didactic GmbH sensor-cassy. The detector was also housed in a thick lead jacket to reduce the radiation background as low possible, the distance between detector and source was 15cm [12]. For each sample and energy, I₀ and I intensities which are without and after attenuation were measured by a NaI (Tl) detector. The photo peak areas have been calculated from the spectrum obtained for each measurement. Background spectra were recorded for the same time period (1000 S) and subtracted from each spectrum. The typical spectra of the radioactive sources ¹³⁷Cs measured in this work are shown in Fig. 2b.

Results and Discussion

- Mass attenuation coefficient

The calculated values of mass attenuation coefficient for PbO+SiO₂ and Fe₂O₃+SiO₂ glass samples at photon energies varying from 1keV to 100MeV are shown in Figs.3 and 4 respectively. Mass attenuation coefficient for Pb+SiO₂ and Fe₂O₃ +SiO₂ glass samples sharply decrease with increase of the photon energy from 0.001 to 0.1 MeV with a peak due to photoelectric effect around the K,L,M-absorption edges of the lead, K absorption edge of the silicon and K absorption edge of the iron. At photon energy from 0.1 to 5 MeV the variation is slightly decreased with the increase of the photon energy. While, at photon energies from 5 to 100 MeV, inconsiderable the increase of the mass attenuation coefficient values have been observed with increase of the photon energy, this may be due to the dominance of pair production in this energy region. Figs.3 and 4 can also be seen the mass attenuation coefficients of PbO+SiO₂ glass samples are higher than that of Fe₂O₃+SiO₂ glasses. This means that there is more photon absorption in the PbO+SiO₂ glass than in the Fe₂O₃+SiO₂ glass. Mass attenuation coefficient increases (Fig.3) with the increase in weight fraction of PbO, this can be attributed to increasing values of Pb which has higher atomic number as compared to other elements. It is estimated that 75 wt% PbO+25wt% SiO₂ glass sample represents best glass sample in terms of gamma ray shielding applications. Figs. 5 and 6 show plot of ln (I₀/I) versus thickness samples at the 0.662 and 1.28 MeV gamma ray beams using this graphs, The slope of the graphs gives the value of the linear attenuation coefficient (μ) of glass samples at that particular energy. Tables 2 and 3 give the experimental and theoretical values of mass attenuation coefficients (μ_m) for glass samples. The comparison of their measurements with the theoretical values is done by calculating the relative deviation

(RD). We found the deviation mostly below 5%. In general, the experimental values are in a good agreement with the theoretical values.

-Effective Atomic Numbers

The variation of effective atomic numbers for selected glass samples (Z_{eff}) with photon energy were calculated from Eq.(5) and plotted in Fig.7. The effective atomic numbers of PbO+SiO₂ glasses are greater than Fe₂O₃ +SiO₂ glasses'. In addition, Z_{eff} increases with the increase of Fe₂O₃ and PbO concentrations. The Z_{eff} values for PbO+SiO₂ glasses show a broad peak and a maximum value at 0.01 MeV and minima at 1 MeV. The variation of Z_{eff} with energy may be attributed to the relative domination of the partial processes, viz. photoelectric effect, coherent scattering, incoherent scattering and pair production. At low energies the photoelectric effect is dominant and hence Z_{eff} for the photon absorption is mainly described by Z_{eff} for this partial process. Similarly, at higher energies the contribution due to scattering and pair production process will be more in comparison with photoelectric effect and this will have its effect on Z_{eff} for photon absorption. Hence at low energies (up to 0.1 MeV), where photoelectric effect dominates, Z_{eff} value is more and at intermediate energies (0.1- 1 MeV), where the scattering process dominate, Z_{eff} value is less. Finally, at higher energies (more than 1.022MeV) the pair production process dominates Z_{eff} value increase with the increase of photon energy. Therefore, Z_{eff} for photon energy absorption varies from a higher value at lower energies to a lower value at intermediate energies with a peak due to photoelectric effect around the K-absorption edge of the lead (0.088MeV).

-Half value layer

The half value layers (HVL) of Fe₂O₃+SiO₂ and PbO+SiO₂ glass samples at photon energies varying from 1keV to 100MeV are shown in Figs.8 and 9 respectively with standard commercial window glass taken from literature [13]. It is found that the PbO+SiO₂ glasses have better shielding properties (gives low HVL at all energies) than commercial window glass and Fe₂O₃+SiO₂, reflecting the advantage of lead component in radiation shielding glass. The HVL values for the PbO+SiO₂, Fe₂O₃+SiO₂ glass samples and standard commercial window glass were also experimentally determined at gamma energies 0.662 and 1.28 MeV as shown in Tables 4 and 5 in addition that plotted in Fig.10. It is evident that the 50wt.%Fe₂O₃+50 wt.%SiO₂ and 25wt.%PbO+75wt.%SiO₂ glass samples have the same HVL values because they have similar effective atomic numbers at particular energies (see Fig.7). The composite of 75 wt.% PbO+25wt.% SiO₂ glass sample have low HVL values than those other selected glass samples, this can be attributed to increasing values of Pb which has higher atomic number as compared to other elements.

Conclusions

From the measurement and calculation of gamma attenuation coefficients in Fe₂O₃ and PbO reinforced in silicate glasses, it can be concluded:

1. The mass attenuation coefficients of the PbO+ SiO₂ glasses are higher than that of the Fe₂O₃+ SiO₂ glasses at the same incident photon energy. The 75 wt.% PbO +25 wt.% SiO₂ proved to be more efficient for gamma rays attenuation.
2. The results show that the effective atomic numbers of PbO+ SiO₂ glasses are larger than that of Fe₂O₃+ SiO₂ glasses and both are greater than SiO₂ glass sample. Moreover, the effective atomic numbers increase with the increase of fraction weight of Fe₂O₃ and PbO.
3. In the case of Fe₂O₃+ SiO₂ glasses, there is only a little change in the half value layer with increase of Fe₂O₃ concentration. We found that the HVL in 50wt.%Fe₂O₃+50 wt.%SiO₂ and 25wt.%PbO+75wt.%SiO₂ glasses have equivalent values.
- 4.The theoretical values of gamma ray attenuation coefficients were generally in good agreement with experimental obtained values.

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Table No. (1): Chemical composition and density of glass samples

Sample number	Composition (wt%)			Density (g/cm ³)
	PbO	Fe ₂ O ₃	SiO ₂	
1	-	-	100	1.9400 ± 0.0132
2	25	-	75	2.7215 ± 0.0120
3	50	-	50	3.8351 ± 0.0157
4	75	-	25	5.0219 ± 0.0140
5	-	25	75	2.6091 ± 0.0132
6	-	50	50	2.9372 ± 0.0161
7	-	75	25	3.4179 ± 0.0154

Table No. (2): Comparison between mass attenuation coefficients μ_m (cm²/g) for PbO+SiO₂ and Fe₂O₃+SiO₂ glasses at photon energy 0.662 MeV

Fe ₂ O ₃ or PbO (wt.%)	PbO+SiO ₂ glass			Fe ₂ O ₃ +SiO ₂ glass		
	μ_m theory (cm ² /g)	μ_m experiment (cm ² /g)	RD %	μ_m theory (cm ² /g)	μ_m experiment (cm ² /g)	RD %
25	0.08490	0.08352 ± 0.0025	1.63	0.07660	0.07994 ± 0.0022	4.36
50	0.09252	0.09476 ± 0.0022	2.42	0.07594	0.07969 ± 0.0018	4.94
75	0.10010	0.09857 ±	1.53	0.07527	0.07244	3.76

Table No.(3): Comparison between mass attenuation coefficients μ_m (cm²/g) for PbO+SiO₂ and Fe₂O₃+SiO₂ glasses at photon energy 1.28 MeV

Fe ₂ O ₃ or PbO (wt. %)	PbO+SiO ₂ glass			Fe ₂ O ₃ +SiO ₂ glass		
	μ_m theory (cm ² /g)	μ_m experiment (cm ² /g)	RD %	μ_m theory (cm ² /g)	μ_m experiment (cm ² /g)	RD %
25	0.05659	0.05884 ± 0.0021	3.98	0.05566	0.05584 ± 0.0010	0.32
50	0.05694	0.05411 ± 0.0010	4.97	0.05507	0.05417 ± 0.0012	1.63

Table No. (4): Comparison between half value layer HVL (cm) for PbO+SiO₂ and Fe₂O₃+SiO glasses at photon energy 0.662 MeV

Fe ₂ O ₃ or PbO (wt.%)	PbO+SiO ₂ glass			Fe ₂ O ₃ +SiO ₂ glass		
	HVL theory (cm)	HVL experiment (cm)	RD %	HVL theory (cm)	HVL experiment (cm)	RD %
25	3.000	3.050 ± 0.089	1.67	3.468	3.323 ± 0.089	4.18
50	1.953	1.907 ± 0.043	2.36	3.108	2.961 ± 0.065	4.30
75	1.390	1.400 ± 0.056	0.72	2.694	2.799 ± 0.108	3.90

Table No.5: Comparison between half value layer HVL (cm) for PbO+SiO₂ and Fe₂O₃ +SiO₂ glasses at photon energy 1.28 MeV

Fe ₂ O ₃ or PbO (wt.%)	PbO+SiO ₂ glass			Fe ₂ O ₃ +SiO ₂ glass		
	HVL theory (cm)	HVL experiment (cm)	RD %	HVL theory (cm)	HVL experiment (cm)	RD %
25	4.501	4.329 ± 0.149	3.82	4.773	4.758 ± 0.084	0.31
50	3.174	3.340 ± 0.061	5.23	4.285	4.356 ± 0.094	1.66
75	2.409	2.453 ± 0.043	1.83	3.722	3.796 ± 0.136	1.99

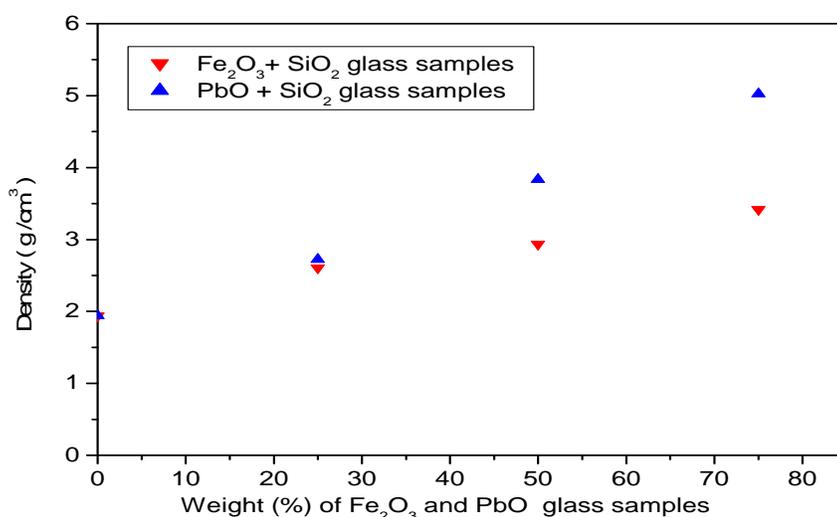


Fig. 1. Densities of the Fe₂O₃+SiO₂ and the PbO+SiO₂ glass systems.

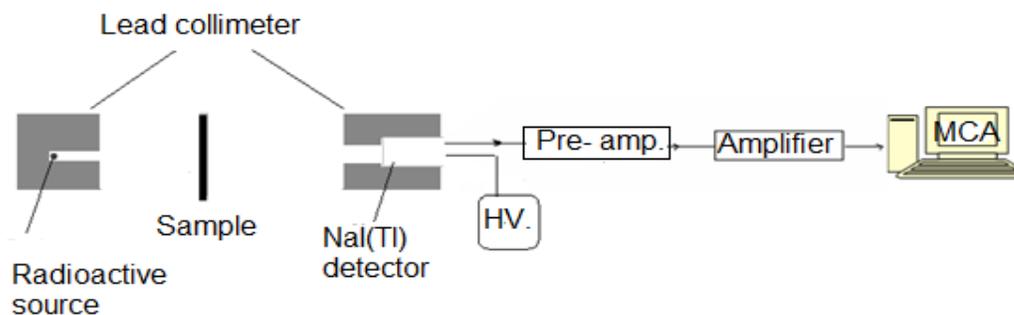


Figure No.(2a): Experimental setup to determine mass attenuation coefficient.

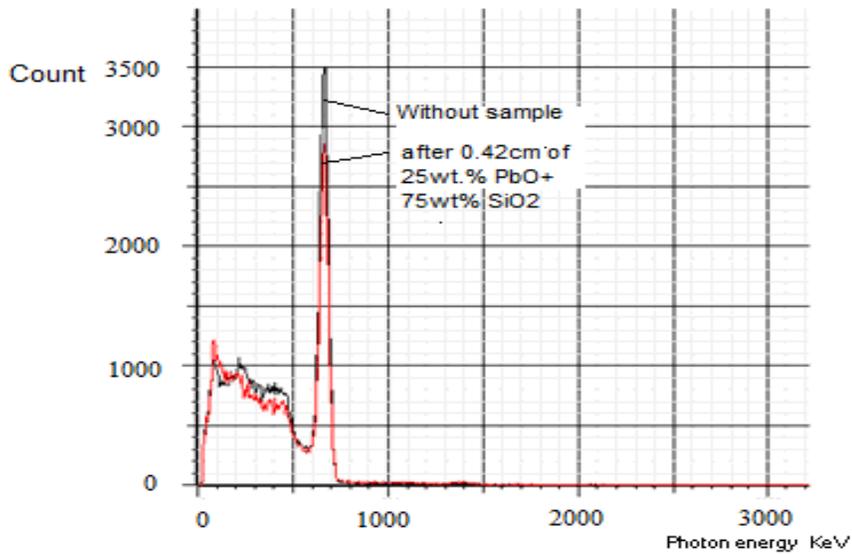


Fig. 2b. Energy spectra of gamma rays emitted from Cs-137 without sample and after 0.42cm of 25wt % +75wt.% SiO₂ glass sample.

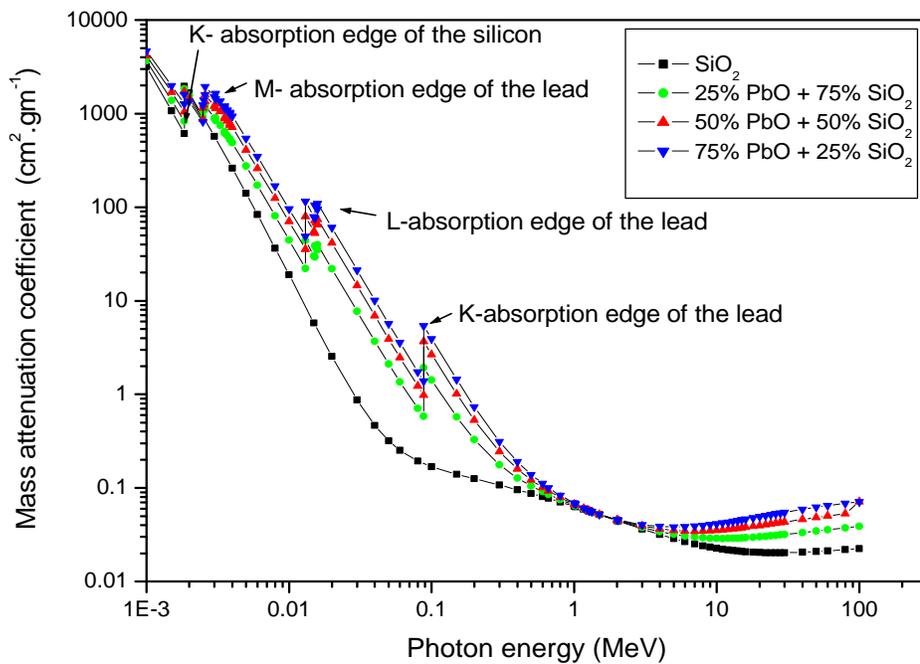


Figure 3. Mass attenuation coefficient as a function of photon energy (1keV to 100 MeV) for SiO₂ and PbO-SiO₂ glasses

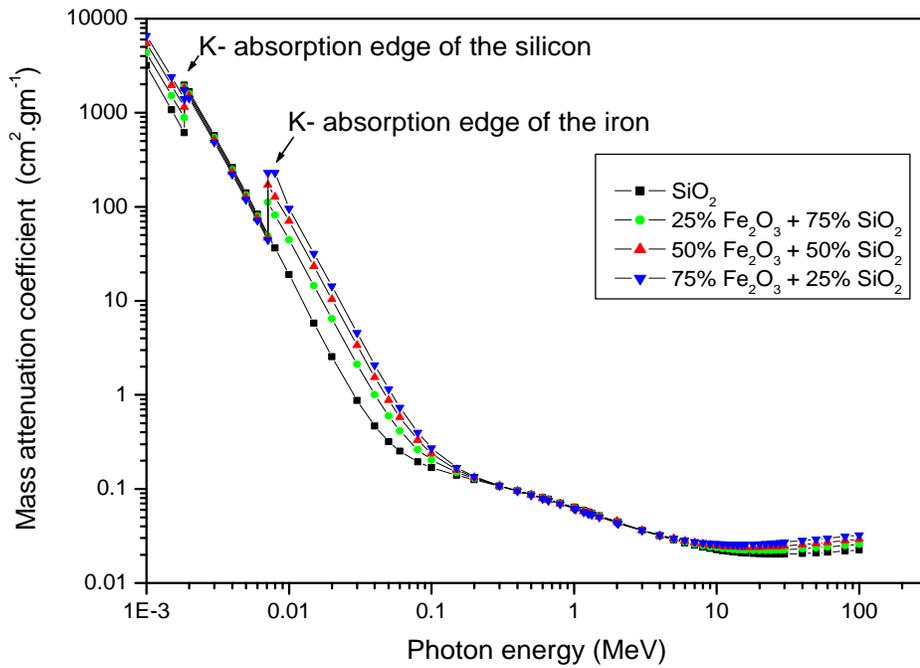


Figure 4. Mass attenuation coefficient as a function of photon energy (1keV to 100 MeV) for SiO_2 and Fe_2O_3 - SiO_2 glasses

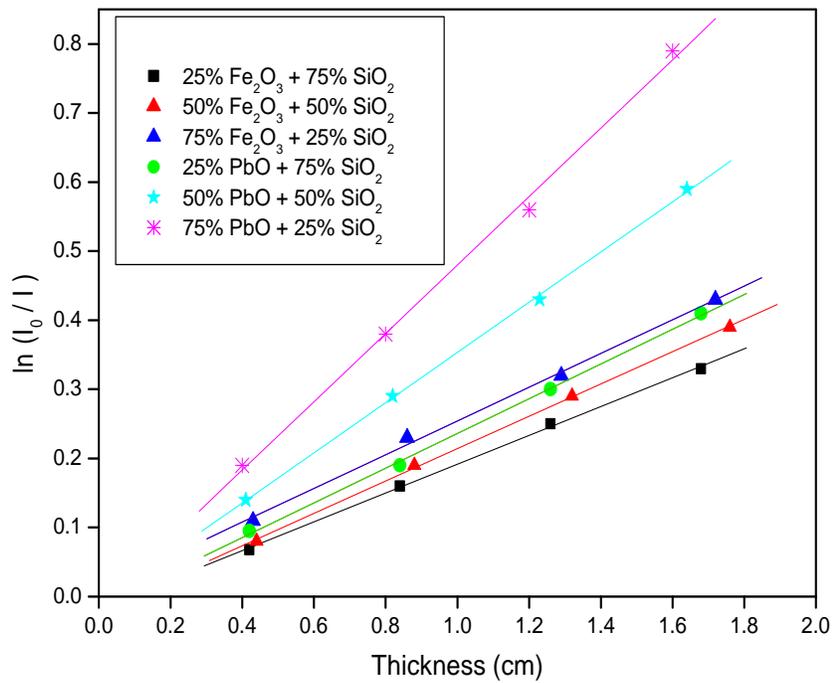


Figure 5. $\ln(I_0/I)$ versus thickness for selected samples at 0.662 MeV.

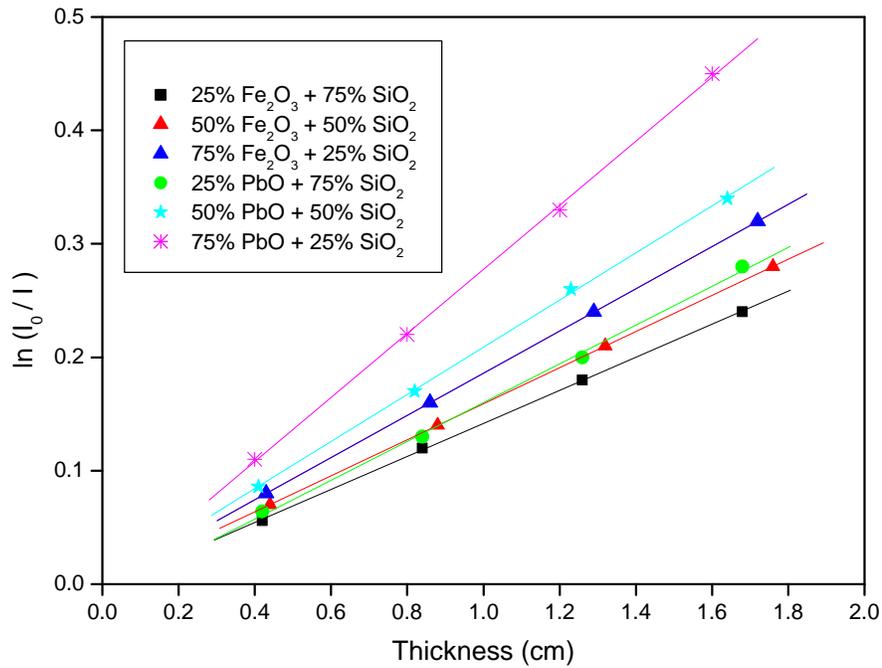


Figure 6. $\ln(I_0/I)$ versus thickness for selected samples at 1.28 MeV.

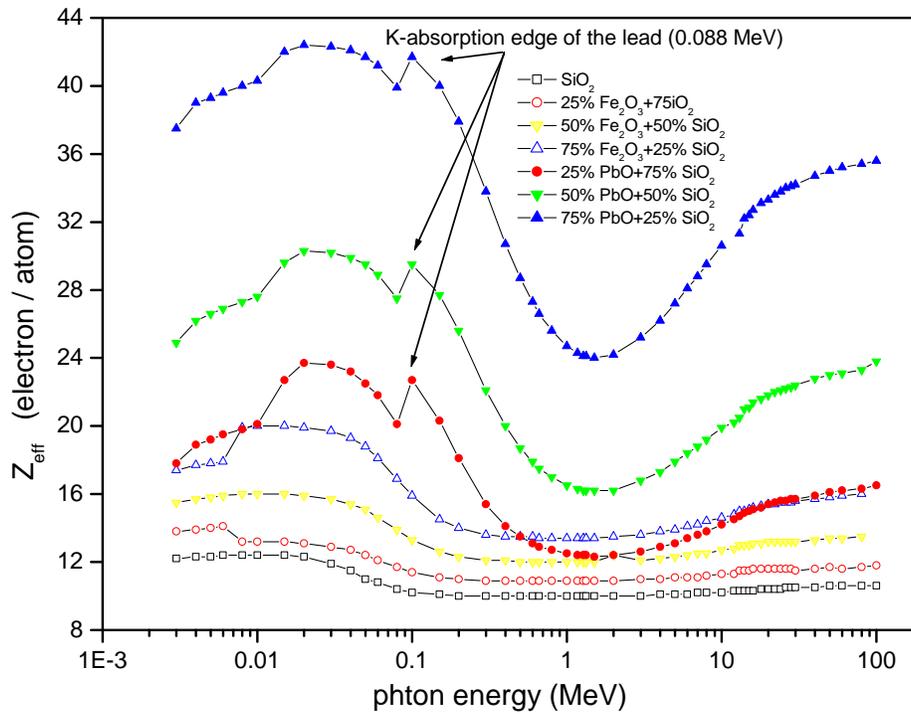


Figure 7. Energy dependence of effective atomic number Z_{eff} for total photon interaction.

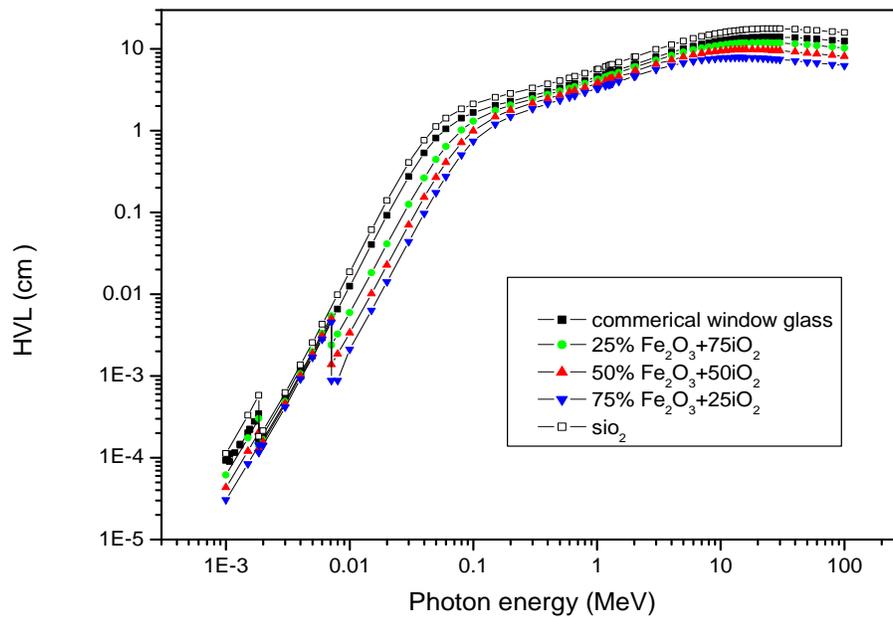


Figure 8. Half value layer as a function of photon energy (1keV-100MeV) in the Fe_2O_3 - SiO_2 glass and commercial glass.

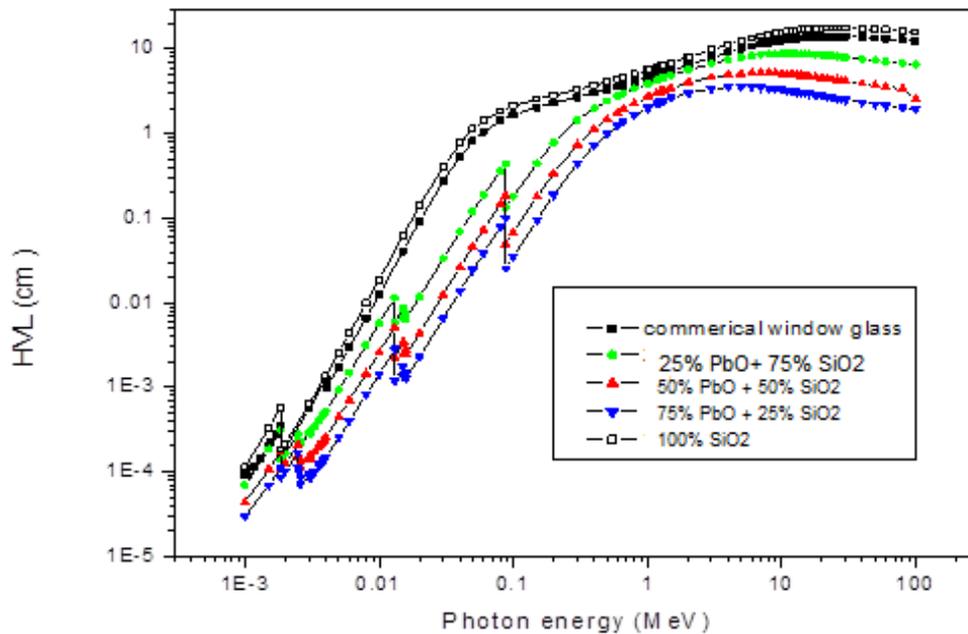


Figure 9. Half value layer as a function of photon energy (1keV-100MeV) in the PbO - SiO_2 glass and commercial glass.

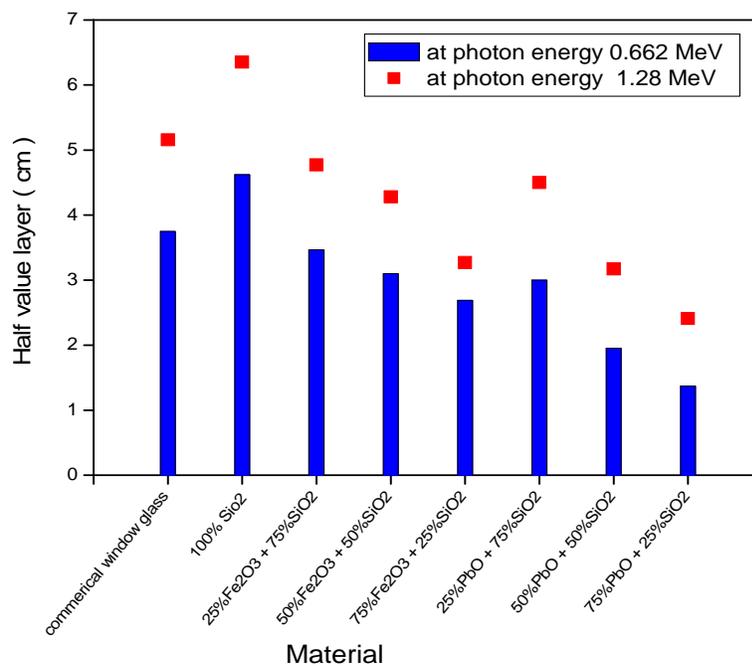


Fig. 10. Half value layer (HVL) of $\text{Fe}_2\text{O}_3+\text{SiO}_2$ glasses compared with those of $\text{PbO}+\text{SiO}_2$ glasses and commercial window glass at 0.662 and 1.28 MeV.

معاملات توهين اشعة كاما في الزجاج المدعم بأوكسيد الرصاص و أوكسيد الحديد كنوافذ للتدريع الأشعاعي

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استلم البحث : 1 شباط 2014 قبل البحث : 2 حزيران 2014

الخلاصة

تم في هذا البحث حساب معامل التوهين الكتلي والعدد الذري المؤثر و سمك النصف لنماذج زجاجية مكونة من اوكسيد السليكون مادة اساس مدعمة باوكسيد الحديد الثلاثي, واوكسيد الرصاص بتراكيز وزنية مختلفة (25, 50, 75) عند مدى واسع من طاقة الفوتون تراوحت من 1keV الى 100MeV باستخدام البرنامج العالمي (XCOM) . كذلك حسب معامل التوهين الكتلي وسمك النصف للنماذج عمليا باستخدام حزمة ضيقة من اشعة كاما عند الطاقات 0.662 MeV و 1.28MeV المنبعثة من النظائر المشعة ^{137}Cs و ^{22}Na على التوالي. استعمل لهذا الغرض الكاشف الومضي "2"x2" NaI(Tl) فكانت هذه القيم موافقة للقيم النظرية. فضلا عن ذلك قورنت النتائج أيضا مع زجاج النوافذ التجاري. تشير نتائج العدد الذري المؤثر وسمك النصف أن زجاج $\text{PbO} + \text{SiO}_2$ أفضل توهينا لأشعة كاما من الزجاج $\text{Fe}_2\text{O}_3 + \text{SiO}_2$, و زجاج النوافذ التجاري وهذا يبين استعمال الزجاج $\text{PbO} + \text{SiO}_2$ للحماية من أشعة كاما بدل الزجاج $\text{Fe}_2\text{O}_3 + \text{SiO}_2$ و زجاج النوافذ التجاري في هذا المدى من الطاقة.

الكلمات المفتاحية: أشعة كاما ,معامل التوهين الكتلي, معامل التوهين الخطي, العدد الذري المؤثر, سمك النصف, الزجاج, مواد التدريع.