
Araştırma Makalesi / Research Article

Gamma-ray Shielding Properties of Lithium Borate Glass Doped with Colemanit Mineral

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Abstract

In this study, $(\text{Li}_2\text{B}_4\text{O}_7)_{(100-x)}-(\text{Colemanite})_x$ glass systems (where $x=10, 20, 30$ and 40 wt %) were fabricated via melt quenching technique. The radiation shielding parameters of produced glasses such as mass attenuation coefficient (μ_m), effective atomic number (Z_{eff}), electron density (N_{el}), half value layer (HVL) and mean free path (MFP) were measured experimentally for 81, 276, 302, 356, and 383 keV gamma ray energies with Hp-Ge detector. Also, WinXCom software was employed for theoretical calculation above radiation shielding parameters of glasses for 1 keV to 10^5 MeV energy region. It was seen that the experimental and theoretical results are good agreement with each other. The obtained results revealed that when percentage of colemanite mineral has been increased in the glass system, μ_m , Z_{eff} and N_{el} values increase. Furthermore, it was observed that the values of HVL and MFP, in contrary, decreases with increasing colemanite mineral percent. In specific, among the investigated glasses, lithium borate glass with 40 percentage colemanite mineral has the highest value of μ_m , Z_{eff} and N_{el} , however, it has the lowest HVL and MFP. Therefore, lithium borate glass with 40 percentage colemanite mineral has given the best results for gamma radiation shielding purpose among the investigated glass systems in this work.

Keywords: Colemanit, lithium borate, mass attenuation coefficient, effective atomic number, half value layer, mean free path

Kolemanit Minerali ile Katkılanmış Lityum Borat Camların Gama-ışını Zırh Özellikleri

Öz

Bu çalışmada, $(\text{Li}_2\text{B}_4\text{O}_7)_{(100-x)} - (\text{Kolemanit})_x$ cam sistemler (burada $x = 10, 20, 30$ ve % 40 ağırlık) erime söndürme tekniği ile üretildi. Üretilen camların, kütle soğurma katsayısı (μ_m), etkin atom numarası (Z_{eff}), elektron yoğunluğu (N_{el}), yarı değer kalınlıkları (HVL) ve ortalama serbest yol (MFP) gibi radyasyon zırh parametreleri 81, 276, 302, 356 ve 383 keV gama enerjilerinde deneysel olarak Hp-Ge dedektörü ile ölçüldü. Ayrıca, camların bu radyasyon zırh parametrelerinin 1 keV- 10^5 MeV enerji aralığında teorik hesaplamaları için WinXCom programı kullanıldı. Deneysel ve teorik sonuçların birbirleri ile iyi uyum içinde olduğu görüldü. Elde edilen sonuçlar, cam sistemindeki kolemanit mineralinin yüzdesi arttığında, μ_m , Z_{eff} ve N_{el} değerlerinin de arttığını ortaya koydu. Ayrıca, bunun aksine, kolemanit yüzdesinin artmasıyla HVL and MFP değerlerinin azaldığı gözlemlendi. Spesifik olarak, incelenen camlar arasında yüzde 40 kolemanit mineral içeriğine sahip lityum borat cam en yüksek μ_m , Z_{eff} ve N_{el} değerine sahip, bununla birlikte en düşük HVL ve MFP'ye sahiptir. Bu nedenle, bu çalışmada, incelenen cam sistemleri arasında %40 kolemanit minerali içeren lityum borat cam gama radyasyonu zırh özellikleri için en iyi sonucu vermiştir.

Anahtar Kelimeler: Kolemanit, lityum borat, kütle soğurma katsayısı, etkin atom numarası, yarı değer kalınlığı, ortalama serbest yol

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1. Introduction

X-rays and gamma rays emitted by radiation sources, which are used in medical treatment, nuclear power plants, agriculture, engineering, radiation biophysics, space technology, nuclear diagnostics etc..., are dangerous to human health and precision laboratory instruments [1]. For instance, direct exposure to gamma radiation with ionizing property can cause organ failure, cancer, skin rash, and genetic damage. Even if human being are exposed to high dose gamma radiation, the result could be death [2]. To minimize harmful effects of gamma and X-rays on human health and instruments, shielding materials have been used. The ideal materials for gamma rays shielding should have some desirable properties such as elements with high atomic number, economically cheap, abundant materials on earth, flexible for applying to any design etc...[3]. Many studies on gamma radiation shielding have been performed by researchers, using different materials such as concrete [4,-7], alloy [8-10] polymer [11,12]. During the last few years, many researchers have paid attention on glass type shielding materials due to transparent property, easy shaped, easy transportation and having properties for attenuation of neutron and photons [13]. Nowadays, many researches related with different type of glass such as Pb-Based Silicate, Borate, and Phosphate glasses [14] heavy metal oxide glasses [15,16], heavy metal fluoride based tellurite-rich glasses [17], optical glasses [18], bismuth borate glasses [19] have been carried out to determine shielding properties. Lithium borate glasses (LBGs) could be good candidate for shielding purpose among the other type of glasses due to high radiation resistance, linear dose dependence in a wide range of doses and high transparency in the spectral range from vacuum ultraviolet to the far infrared region [20].

Colemanite ($2\text{CaO} \cdot 3\text{B}_2\text{O}_3 \cdot 5\text{H}_2\text{O}$) is calcium borate mineral. It is one of the fairly used aggregate added in radiation shielding concrete because of its the least soluble property as a natural borates [21].

After very intense literature search, I could not come cross any theoretical or experimental studies related with investigation of radiation shielding properties of lithium borate glasses doped with colemanite mineral. Therefore, this study will be guide to researchers who performs on the LBGs systems.

The main parameters of gamma shielding interaction with materials are counted as a mass attenuation coefficient, effective atomic number, half-value layer, and mean free. In this study, these parameters for lithium borate glasses doped without and with colemanite mineral in various percentage were determined not only with experimental method, but also with WinXCom software in the energy range 1keV-100GeV.

2. Material and Method

2.1. Theoretical Calculations

2.1.1. Mass attenuation coefficient

If monochromatic-ray pass through matter, a part of them is absorbed. The decrease in radiation intensity is expressed by an exponential equation containing parameters related to the given case [22]:

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

where I is the intensity of transmitting radiation; I_0 is the incident radiation intensity; μ represents the linear attenuation coefficient, cm^{-1} ; x stand for the material thickness, cm.

The mass attenuation coefficient, $\mu_m = \frac{\mu}{\rho}$, is related to unit density (ρ , g/cm^3); then Equation 1 have the following form:

$$I = I_0 e^{\mu_m \rho x} \quad (2)$$

If target consist of mixture and compound, the mass attenuation coefficient is calculated via WinXCom software according to mixture law [23].

$$\mu_m = \sum_i P_i \mu_{m,i} \quad (3)$$

where P_i and $\mu_{m,i}$ are the weight fraction and mass attenuation coefficient, respectively, of the constituent element i^{th} .

$$\sum_i P_i = 1 \quad (4)$$

2.1.2. Effective atomic number and electron density

Effective atomic number, Z_{eff} , represents interaction between gamma-ray and absorbent composed from various elements Effective atomic number is written in the form following [24] :

$$Z_{\text{eff}} = \frac{\sum_i f_i A_i \mu_{m,i}}{\sum_j f_j \frac{A_j}{Z_j} \mu_{m,j}} \quad (5)$$

where f_i is the fraction by mole of each constituent element, providing that $\sum_i f_i = 1$, A_i is the atomic weight, Z_j is the atomic number and $\mu_{m,i}$ is the mass attenuation coefficient of i^{th} element.

Effective electron density, N_{el} (electrons/gram), describes the number of electrons per unit mass of the interacting materials and it is given with following formula [25].

$$N_{el} = N_A \frac{n Z_{\text{eff}}}{\sum_i n_i A_i} = N_A \frac{Z_{\text{eff}}}{A} \quad (6)$$

here, A is the mean atomic mass and N_A is Avogadro constant.

2.1.3. Half value layer and mean free path

Half value layer (HVL) is the thickness of a given material which decreases the intensity of radiation to half of its original value as a follows [26]:

$$HVL = \frac{0.693}{\mu} \quad (7)$$

Mean free path (MFP) is the average distance which a photons travels in medium before an interactions occurs (or between successive interactions) and have been calculated using the following relation [27]:

$$MFP = \frac{1}{\mu} \quad (8)$$

3. Experimental Method

3.1. Preparation of sample

Doping mineral colemanite is mined at Kutahya province in Turkey (Emet, Hisarcık areas) and its chemical content was determined by X-ray diffraction(XRF) [28]. Then, to fabricate pure LBG, the rapid melt quenching technique was employed. In this technique, lithium borate powder was melted within a platinum crucible of an electrical furnace at the temperature of 1000°C for 30 min. Obtained melt was poured in a disc-shaped platinum plate. After that, desired percentage colemanite element was added to lithium borate powder and above procedure was repeated to fabricate aimed glass samples. The prepared samples have 37mm diameter and their thicknesses are 3.6 ± 0.02 mm. The pure lithium borate glass was used as a reference sample and labeled with R. According to added percentage of colemanite mineral, samples were labeled with C1, C2, C3 and C4 for 10%, 20%, 30% and 40%, respectively. Density of glass samples was measured via Archimede's principle (Table1).

3.2. Gamma-ray transmission measurements

In this study, HpGe detector was used to measure photon interaction parameters of lithium borate glass with and without doping minerals. Gamma photons, which are emitted from ^{137}Cs point source, with 60, 112, 204, 320, 464 and 662 keV energies were used to irradiate samples. After irradiation progress, the photo peak density was measured before and after sample was placed between radioactive source and detector (Figure 1). The spectrums were recorded using the computer software MAESTRO (6.08 version) with a multichannel analyzer. The data for every measurements were collected for one hour and the measurement for each sample were repeated 3 times. The mass attenuation coefficient value was calculated after value of I_0 and I were inserted in Equ-1.

The experimental uncertainty of attenuation coefficient measurements for the fabricated glasses has been estimated by relation [29]:

$$\Delta\left(\frac{\mu}{\rho}\right) = \frac{1}{\rho x} \sqrt{\left[\left(\frac{\Delta I_0}{I_0}\right)^2 + \left(\frac{\Delta I}{I}\right)^2 + \ln\left(\frac{I_0}{I}\right)^2 \cdot \left(\frac{\Delta \rho x}{\rho x}\right)^2\right]}$$

where $\Delta \rho x$ denotes the uncertainty in the mass per unit area, ΔI_0 and ΔI are the uncertainty in the intensities I_0 , I , respectively.

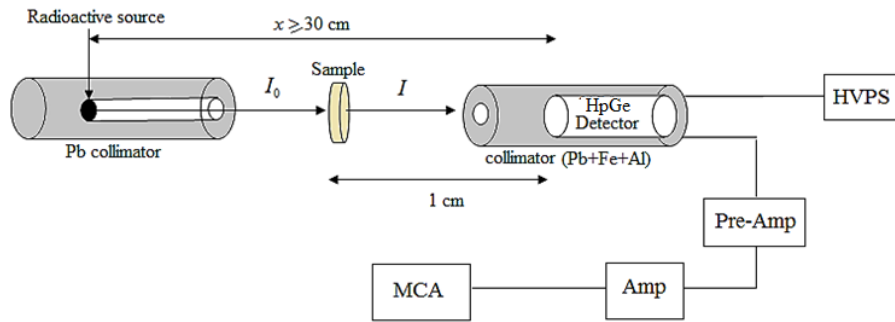


Figure 1. Experimental set up for measurement

4. Result and Discussion

The mass attenuation coefficients (μ_m) values obtained from experimental measurement with 3 % statistical error and theoretical calculation were inserted in Table 1. As seen from Table 1, theoretical and experimental findings are in good agreement with each other in experimental energy limits. Moreover, while values of μ_m decrease with increasing photon energies, they increase with increasing colemanite mineral percentage. Figure 2a demonstrates theoretical μ_m values calculated by WinXCom software versus incident photon energy for 1 keV to 10^5 MeV. In Figure 2a, energy scale could be divided into three energy zones according to the dominant γ -photon interaction: photoelectric effect ($E < 0.05$ MeV), Compton scattering ($0.05 \text{ MeV} < E < 30 \text{ MeV}$) and pair production ($E > 30 \text{ MeV}$). In the photoelectric zone (Figure 2b), the value of μ_m decreases sharply with increasing energy for the investigated LBGs. This case can be explained by the fact that the microscopic cross section is inversely correlated with the incoming photon energy $E^{-3.5}$, however, it is directly proportional to the LBGs atomic number, $Z^{4.5}$ for the photoelectric effect. Figure 2c shows how the values of μ_m change with energy in the Compton zone. The values of μ_m slowly decrease and the difference in μ_m values of LBGs is almost zero. The reason for the variation in μ_m with energy depends on the cross section of Compton scattering, which is related linearly to the atomic number (Z). After 30 MeV, corresponding to the pair production region (Figure 2d), μ_m slightly increases and then stays almost constant (for γ -ray energies $> 10^3 \text{ MeV}$). This case is explained by the Z^2 dependence of the pair production process [30]. The highest value of μ_m belongs to the C4 sample, while the R sample has the lowest value.

Table 1. μ_p , Z_{eff} and HVL values of glasses acquired as theoretically and experimentally

	Li ₂ B ₄ O ₇ (Ref. Glass)		Colemanite %10		Colemanite %20		Colemanite %30		Colemanite %40	
	2.210±0.004 g/cm ³		2.254±0.004 g/cm ³		2.271±0.004 g/cm ³		2.288±0.004 g/cm ³		2.306±0.004 g/cm ³	
Energy	μ/ρ (cm ² /g)									
(MeV)	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.
0.081	0.1593	0.1590±0.0047	0.1631	0.1598±0.0041	0.1661	0.162±0.0042	0.1690	0.1684±0.0054	0.1720	0.1752±0.0051
0.276	0.1068	0.1065±0.0031	0.1070	0.1112±0.0035	0.1082	0.110±0.0035	0.1091	0.1121±0.0033	0.1092	0.1108±0.0034
0.302	0.1032	0.1030±0.0030	0.1042	0.0995±0.0024	0.1041	0.106±0.0034	0.1055	0.1092±0.0037	0.1057	0.1084±0.0032
0.356	0.0971	0.0973±0.0029	0.0987	0.0957±0.0022	0.0981	0.101±0.0032	0.0994	0.1128±0.0035	0.0999	0.1032±0.0032
0.383	0.0942	0.0940±0.0028	0.0953	0.0924±0.0027	0.0949	0.098±0.0024	0.0961	0.1005±0.0034	0.0966	0.0965±0.0024
Energy	Z_{eff} (electron/atoms)									
(MeV)	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.
0.081	6.7591	6.7480±0.2024	6.9881	6.8178±0.2041	7.1867	7.0134±0.2104	7.3854	7.342±0.2202	7.7849	7.9205±0.2370
0.276	6.7478	6.7287±0.2018	6.9413	7.2012±0.2162	7.1906	7.3232±0.2199	7.4411	7.646±0.2291	7.8890	7.9472±0.2387
0.302	6.7538	6.7378±0.2021	6.9869	6.6506±0.1995	7.1704	7.3087±0.2195	7.4247	7.707±0.2312	7.8885	8.0834±0.2426
0.356	6.7534	6.7695±0.2030	7.0034	6.7899±0.2033	7.1892	7.4095±0.2227	7.4506	8.428±0.2524	7.8902	8.2060±0.2469
0.383	6.7541	6.7364±0.2020	6.9938	6.7735±0.2032	7.1797	7.4067±0.2228	7.4434	7.753±0.2327	7.8897	7.9555±0.2387
Energy	HVL(cm)									
(MeV)	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.
0.081	1.9687	1.9725±0.0594	1.8852	1.9340±0.1301	1.8372	1.8842±0.127	1.7925	1.8032±0.1234	1.7470	1.7174±0.1182
0.276	2.9364	2.9447±0.0882	2.8743	2.7702±0.1875	2.8205	2.7745±0.189	2.7764	2.7045±0.1856	2.7521	2.7325±0.1894
0.302	3.0394	3.0447±0.0915	2.9514	3.1065±0.2104	2.9117	2.8790±0.196	2.8719	2.7795±0.1901	2.8600	2.7832±0.1925
0.356	3.2307	3.2235±0.0971	3.1385	3.2368±0.2182	3.1118	3.0212±0.205	3.0474	2.7043±0.1856	3.0368	2.9185±0.2017
0.383	3.3301	3.3364±0.1001	3.2368	3.3425±0.2251	3.2157	3.1144±0.212	3.1529	3.0296±0.2074	3.1272	3.1309±0.2145
Energy	$N_{eq} \times 10^{23}$ (electron/gram)									
(MeV)	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.
0.081	2.9214	2.9178±0.0875	2.9392	2.8673±0.0864	2.9434	2.8721±0.0862	2.9481	2.931±0.087	2.9203	2.9715±0.0892
0.276	2.9170	2.9081±0.0873	2.9197	3.0285±0.0906	2.9447	2.9993±0.0895	2.9704	3.052±0.091	2.9598	2.9817±0.0894
0.302	2.9191	2.9126±0.0872	2.9384	2.7978±0.0832	2.9367	2.9931±0.0893	2.9649	3.077±0.092	2.9595	3.0325±0.0901
0.356	2.9198	2.9265±0.0875	2.9454	2.8555±0.0851	2.9442	3.0349±0.0917	2.9747	3.364±0.100	2.9597	3.0788±0.0928
0.383	2.9193	2.9127±0.0874	2.9416	2.8482±0.0851	2.9406	3.0332±0.0904	2.9716	3.095±0.092	2.9599	2.9845±0.0898
Energy	MFP(cm)									
(MeV)	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.	Theo.	Expt.
0.081	2.8409	2.8460±0.0856	2.7203	2.7901±0.0845	2.6511	2.7185±0.0783	2.5866	2.6025±0.074	2.5214	2.4780±0.0742
0.276	4.2372	4.2498±0.1272	4.1476	3.9979±0.1205	4.0700	4.0032±0.1207	4.0112	3.9025±0.116	3.9714	3.9425±0.1189
0.302	4.3859	4.3937±0.1325	4.2558	4.4812±0.1346	4.2301	4.1545±0.1256	4.1442	4.0102±0.129	4.1034	4.0152±0.1209
0.356	4.6620	4.6507±0.1403	4.4964	4.6704±0.1409	4.4903	4.3607±0.1312	4.3975	3.9027±0.113	4.3426	4.2105±0.1264
0.383	4.8053	4.814±0.1443	4.6554	4.8221±0.1459	4.6403	4.4938±0.1359	4.5495	4.3717±0.132	4.4903	4.5170±0.1367

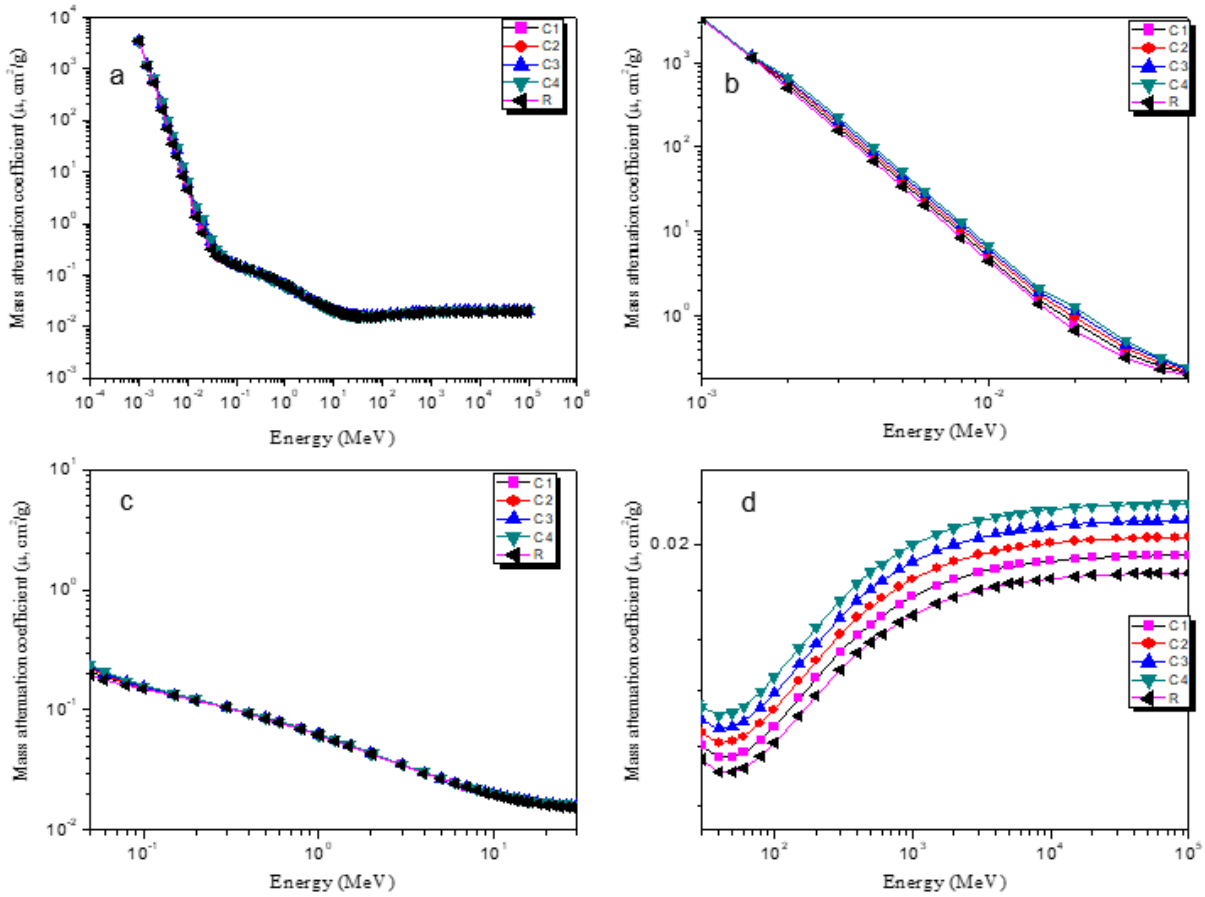


Figure 2. Mass attenuation coefficients for the selected glasses in (a) the total energy region ($1\text{KeV}<E<10^5$ MeV) and (b) the low energy region $E>0.05$ MeV (c) the energy region 0.05 MeV $< E < 30$ MeV (d) the energy region $E > 30$ MeV

The Table 1, demonstrate Z_{eff} ve N_{el} values measured and calculated at selected energy values. As seen from tables, they are in good harmony with each other. Also, Figure 3 - 4 present Z_{eff} ve N_{el} for incident photon energy ($1\text{keV} \leq E \leq 10^5$ MeV). R, C1, C2, C3 and C4 samples have same characteristic behavior for Z_{eff} (Figure3). In low energy region ($1\text{keV} - 0.015$ MeV), Z_{eff} and N_{el} have the highest values (Figure 3-4) for with and without doping minerals of LBG. In specific, the value of Z_{eff} ve N_{el} sharply decrease with incident γ -ray in the range of 0.015 MeV and 0.02 MeV due to photoelectric effect . After 0.02 MeV, they start rise till 0.3 MeV and stay constant for rest of incident gamma-ray energy. Moreover, the value of Z_{eff} increases with increasing percentage of colemanite in system. Same behavior is also observed for N_{el} due to close relation with Z_{eff} . These result also supports to Hine findings [31].

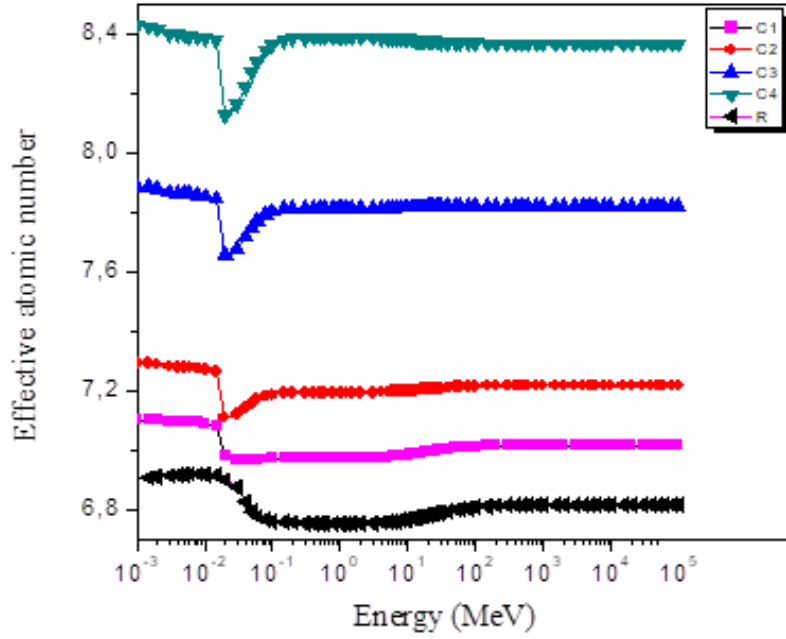


Figure 3. Variation of effective atomic numbers against photon energy of lithium borate glasses

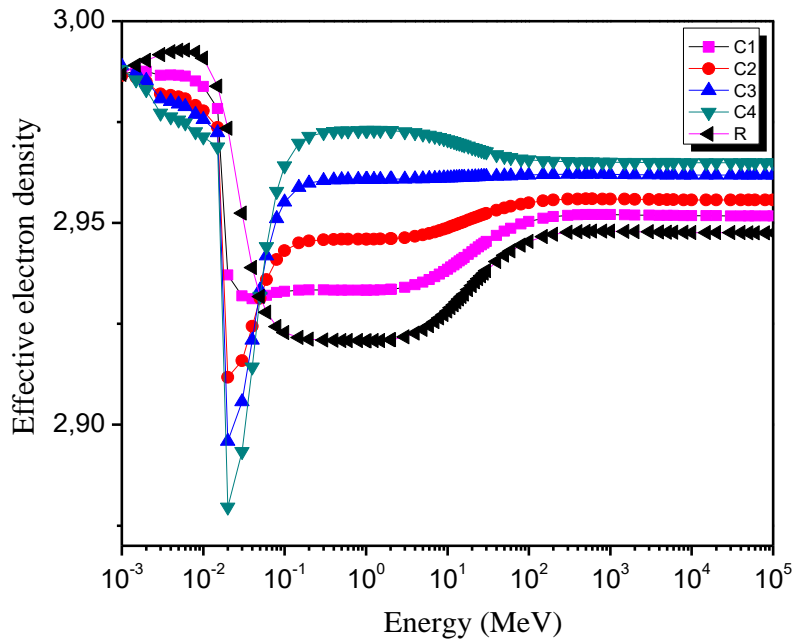


Figure 4. Variation of electron density against photon energy of lithium borate glasses

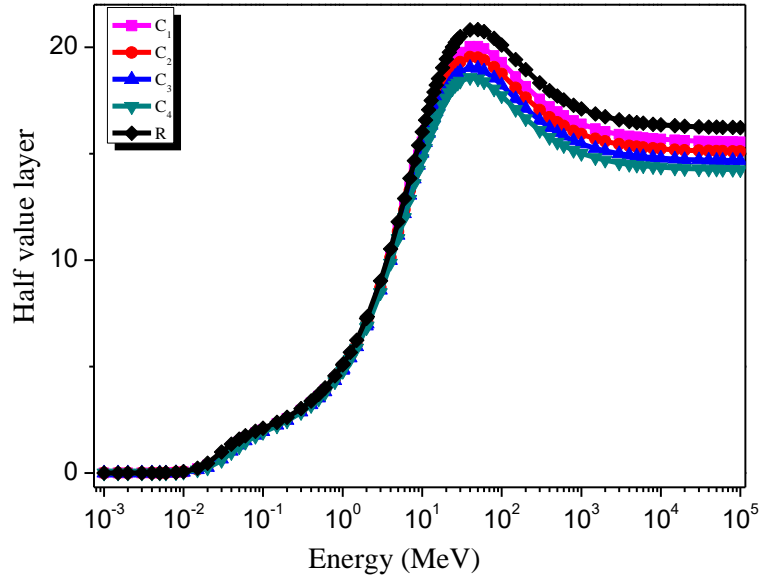


Figure 5. Half value layer (HVL) against photon energy of lithium borate glasses

Half value thicknesses (HVL) and mean free path (MFP) of all samples are also calculated and inserted in Table 1 for the same energy values. In experimental set up limit, the obtained values of HVL and MFP from theoretical calculation agree fairly well with experimental results. Also, Figures 5 and 6 represents HVL and MFP versus incident gamma energies, respectively. As seen from Figure 5 and 6, HVL and MFP have the lowest value and independent from colemanite percentage and incident gamma ray energy in the low energy region ($< 0.01 \text{ MeV}$). In intermediate region ($0.01 \text{ MeV} < E < 30 \text{ MeV}$), the values of HVL and MFP rise sharply. Moving further the high energy region, the HVL and MFP values gets smoother and it stays constant after 2000 MeV. One more detail, while the highest values of HVL and MFP belong to R sample, C4 sample has the lowest values for both parameters which indicates that C4 sample has high shielding capacity.

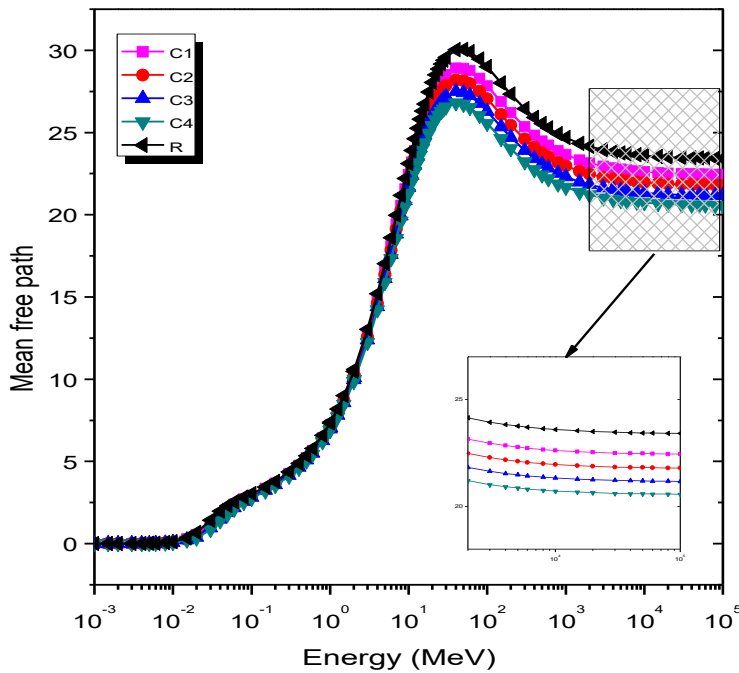


Figure 6. Mean free path (mfp) against photon energy of lithium borate glasses

5. Conclusion

In present work, pure and various percentage colemanite doped lithium borate glasses were fabricated. Using both experimental and theoretical methods, photon interaction parameters such as mass attenuation coefficient (μ_m), effective atomic number (Z_{eff}), electron density (N_{el}), half value layer (HVL) and mean free path (MFP) of LBGs were measured and calculated, respectively. It was observed that the obtained theoretical and measured experimental results verify each other in the experimental energy region limit. Increasing percentage of colemanite mineral in glass systems results to higher mass attenuation coefficient value. On the contrary, the value of mass attenuation coefficient decreases with increasing energy. The μ_m gets higher value in the low energy region where photoelectric phenomena dominates and it has smaller value in the middle energy region where Compton scattering is main process. Among the fabricated glass system, LBG doped with 40 % colemanite mineral has the highest μ_m value. The parameters Z_{eff} and N_{el} are strictly related to photon energy and the percentage rate of colemanite mineral in LBGs systems. The higher values of both parameters are in the lower energy region where the photoelectric effect dominates. To gain better shielding properties, material should have the lower HVL and MFP value which indicates more interaction between gamma rays and material. According to obtained results, LBG doped with colemanite mineral 40 %, which is transparent, could be considered as a shielding material against γ - radiation used in various technological areas.

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