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Effect of La_2O_3 Additive on the Radiation Shielding Properties of Cobalt-doped Borate Glasses

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ABSTRACT: Radiation protection becomes a necessary action in radiology, Nuclear Medicine, Radiotherapy and other institutions which make use of radiation. The aim of this work is to evaluate the radiation protection features of cobalt-doped borate glass with La_2O_3 additive. The glasses named here as S1, S2, S3, S4 and S5 with 0, 0.5, 1, 1.5 and 2 mol% of La_2O_3 respectively were assessed for their Linear Attenuation Coefficient, LAC, Mass Attenuation Coefficient, MAC, Half value Layer, HVL and Effective Atomic Number Z_{eff}. We found that the glasses have LAC, MAC and Z_{eff} higher than ordinary concrete and Ilmenite concrete, especially S3 which has the concentration of 1 mol% of La_2O_3 and has lowest HVL at lower energies below 0.1 MeV and above 4 MeV than the other samples, and also found that S5 with 2 mol% of La_2O_3 is more advantageous for energy range 0.1 - 4 MeV. We therefore concluded the applicability of these glasses for radiation shielding and that the addition of La_2O_3 increases their radiation protection features.

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Radiation nowadays is very important tool in health care, agriculture, power generation etc. But the harmful effects of radiation must be avoided through the use of shielding materials that can lower the radiation energy to safety level (Alhassan et al., 2023; Sayyed et al., 2020) .Concrete, rocks and some clay blocks are among the most used materials for radiation protection (Issa et al., 2020). However, the limitation of these materials to be used only in some situations and their opaque nature make them irrelevant for some applications (Alhassan et al., 2023). Glasses are the current trending materials to substitute the previous ones with transparent nature and are environmentally friendly in addition to their high radiation attenuation ability (Alhassan et al., 2023). Therefore, research works on both their physical and optical properties and on their radiation attenuation ability are going on. Borate glasses doped with other elements or compounds are showing promising results to achieve this aim and therefore become a good research area in Physics, Chemistry and Engineering. Research carried out by (Sardar Pasha et al., 2019) investigated the influence of Lithium Sulphate (Li_2SO_4) on the properties of Lithium Lead Borate glass doped with Neodymium. The addition of Li_2SO_4 into the glass results in an increase in its transition temperature which indicates the structural stiffness of its network. Studies by (Farouk et al., 2015) on the effect of Neodymium on the optical properties of Bismuth Borate glasses doped with Lead also shows improvement in its optical energy band gap feature. Addition of $NdCl_3$ into Bismuth Borotellurite glasses

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was also studied and found to be improving its energy band gap (Rammah et al., 2019). The effect of adding Antimony-oxide to Sodium Borosilicate glasses was studied by (Zoulfakar et al., 2017) and found it to improve its mass attenuation coefficient, MAC, especially at some energy levels. Borate glasses with fixed ratio of chromium trioxide (CrO_3) and varying concentrations of Yttrium Oxide (Y_2O_3) was also prepared by (Sayyed et al., 2022) in order to study its structural and optical properties as well as its radiation shielding ability. They found that the higher the Y_2O_3 the higher the coefficient of linear attenuation, LAC, and the lower the half value layer, HVL, of the glass and found that the glasses have both good optical and shielding qualities. Studies by (Abdel-Ghany et al., 2020) on the influence of iron oxide (Fe_2O_3) on the optical and the structural properties of some Borate glasses mixed with CaO and Na_2O_3 found that, the increase in Fe_2O_3 results in the decrease in the chemical bond energy and the bond force constant in the glass. Borate glasses doped with Cobalt was studied by (Abul-Magd et al., 2021) in order to determine the effect of the rare-earth oxide, Lanthanum Oxide (La_2O_3) on its properties. The glasses were found optically active and could be utilized for nonlinear optical applications. Lead Borate glass has been recommended for having good radiation shielding ability, thermal stability, and optical applications (Abul-Magd et al., 2021). We, therefore, in the present work, aimed at studying the effect of the La_2O_3 additive on the radiation shielding properties of Cobalt doped borate glasses.

MATERIALS AND METHOD

Theoretical Background: According to Beer's Lambert Law, when radiation of initial intensity I_o passes through a material whose thickness is t and linear attenuation coefficient (LAC) is μ, the intensity will reduce to a level, I, (transmitted radiation) given by (Equation 1) (Şakar et al., 2020).

$$I = I_o e^{-\mu t} \text{ ………………………… (1)}$$

Therefore, the final intensity depends on the LAC and the thickness of the shielding material.

To assess the radiation protection ability of glass, some of its features such as density, linear attenuation coefficient (LAC), mass attenuation coefficient (MAC), half value layer, HVL, and effective atomic number Z_{eff} were measured.

Density: Density is defined as the ratio of mass to volume of a material, as given by (Equation 2).

$$\rho = \frac{m}{V} \text{ ……………………………(2)}$$

Where; ρ is the density, m is the mass and V is the volume.

An alternative technique to determine the density of glass and other solids which have no shape that has definite volumetric formula, is to weigh their masses in air and in another liquid, say l, of known density and then apply Archimedes' Principle, given by (Equation 3).

$$\rho_g = \frac{w_a}{w_a - w_l} \times \rho_l \text{ ………………….. (3)}$$

Where ρ_g is the density of the glass under study, w_a is the mass of the glass in air, w_l is the mass of the glass in the liquid, l, and ρ_l is the density of the liquid (Al-Hadeethi et al., 2019).

Mass Attenuation Coefficient (MAC): Mass Attenuation Coefficient is another important parameter which shows the probability of interaction between radiation and mass per unit area of the material through which it passes (Şakar et al., 2020). Mass attenuation coefficient depends on the density of the material and can be calculated using (Equation 4):

$$\mu_m = \frac{\mu_L}{\rho} \text{ ………………………………… (4)}$$

Where ρ is the density of the material and μ_L is its linear attenuation coefficient (LAC).

Half Value Layer, (HVL): Half Value Layer, (HVL) is defined as the thickness of the material that is capable of reducing the radiation energy or intensity to half of its incident energy or intensity respectively. HVL varies based on the energy variation (Alhassan et al., 2020) and the nature of the radiation. HVL can be calculated using (Equation 5).

$$HVL = \frac{0.693}{\mu_L} \text{ ……………………………(5)}$$

Where μ_L is the Linear Attenuation Coefficient of the material?

Effective Atomic Number, Z_{eff}: Effective Atomic Number of a molecule, Z_{eff}, can be calculated using (Equation 6).

$$Z_{eff} = \frac{\delta_a}{\delta_e} \text{ ……………………………… (6)}$$

Where δ_a and δ_e are the effective atomic cross section and the total electronic cross section given by (Equation 7) and (Equation 8) respectively

$$\delta_a = \frac{1}{N_A}\sum_i f_i A_i\,(\mu_m)_i \;\ldots\ldots\ldots\ldots\ldots\ldots\; (7)$$

$$\delta_e = \frac{1}{N_A}\sum_i \frac{f_i A_i}{Z_i}(\mu_m)_i \;\ldots\ldots\ldots\ldots\ldots\ldots\; (8)$$

Where f_i represents the fractional abundance of element, i, relative to the total number of atoms. Ai, Z_i and $(\mu_m)_i$ represents the atomic weight, mass number and mass attenuation coefficient of the ith element respectively, and N_A is Avogadro's number (Boonin et al., 2020; Singh et al., 2014).

Phy-X/PSD Software: Phy-X/PSD is an online software developed for the purpose of research on radiation shielding and dosimetry (Şakar et al., 2020). The software could calculate the Mass Attenuation Coefficient, μ_m, linear attenuation coefficient (μ_L), Half Value Layer, (HVL), Tenth Value Layer (TVL), Effective Atomic Number Z_{eff}, Effective Atomic Cross Section, δ_a, Total Electronic Cross Section, δ_e, and many other radiation shielding features of a materials. The glass we named in this work as S1, S2, S3, S4 and S5 with chemical compositions and densities given in (Table 1) are fabricated by (Abul-Magd et al., 2021). We made use of Phy-X/PSD to simulate the irradiation of the glasses with gamma ray from Cobalt-60 radionuclide and evaluated the effect of adding La_2O_3 to the radiation shielding features of the glasses.

Table 1: the compositions of the glasses S1, S2, S3, S4 and S5.

Composition	CoO (mol%)	B_2O_3 (mol%)	La_2O_3 (mol%)	PbO (mol%)	ρ (gcm^{-3})
S1	0.3	80.00	0.00	19.7	3.11
S2	0.3	79.50	0.50	19.7	3.18
S3	0.3	79.00	1.00	19.7	3.24
S4	0.3	78.50	1.50	19.7	3.29
S5	0.3	78.00	2.00	19.7	3.36

RESULT AND DISCUSSION

Linear Attenuation Coefficient (LAC): The linear Attenuation Coefficient is a property which describes how a material attenuates photon per unit thickness. The higher the LAC of a material at a particular energy the better its shielding ability. (Figure 1) shows the variation of LAC of S1 - S5 glasses with energy within 0.02 MeV (20 keV) – 0.15 MeV (150keV) and (Figure 2) shows their variation within 0.02 MeV (20 keV) – 15 MeV.

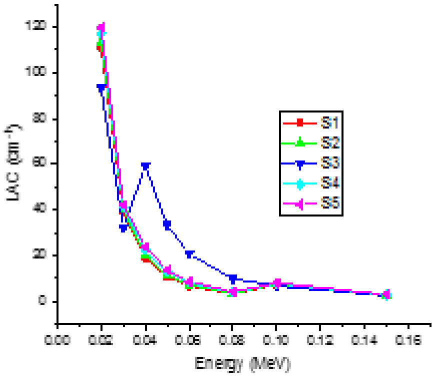

Fig 1: Variation of LAC with photon energies between 0.02 - 0.15 MeV.

Figure 1 shows drastic drop in LAC for all the glasses, followed by an increase to a local maxima, and then a gentle drop. S3 started this pattern by dropping from 178.684 cm^{-1} to 32.018 cm^{-1} between 0.015 MeV (15 keV) and 0.03 MeV (30 keV) and then increased to a maximum of 59.282 cm^{-1} at 0.04 MeV and then gradually decreases as energy increases up to 15 MeV, whereas, for other glasses, the drastic drop is between 0.015 MeV (15 keV) and 0.08 MeV (80 keV) and then they increased to a maximum of 7.330, 7.501, 7.772 and 7.943 cm^{-1} for S1, S2, S4 and S5 respectively. They all then decrease gently as energy increases until 15 MeV.

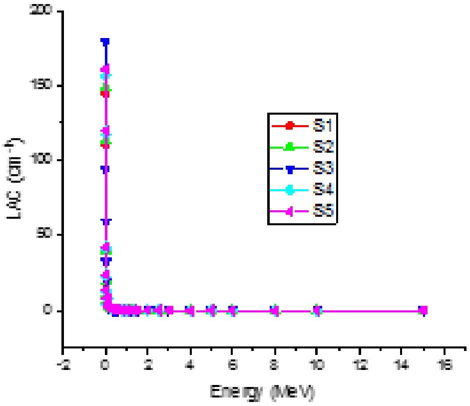

Fig 2: Variation of LAC with photon energies between 0 - 15 MeV.

It can be seen that S3 has the highest value of LAC at 0.015 MeV and attained the highest maximum value after the drastic drop. While for other glasses, the higher the ratio of La_2O_3 additive, the higher the LAC (Figure 1). This phenomenon could be attributed to the Average Molecular Weights (AMW) of the glasses, with S3 has the highest AMW of 425.00 $gmol^{-1}$, followed by S5, S4, S2 and then S1 with AMW of 105.02, 103.73, 101.17 and 99.89 $gmol^{-1}$ respectively.

For this reason, S3 is more suitable for radiation shielding for energies below 0.1 MeV. For energies above 0.1 MeV, S3 has the lowest LAC, while for other glasses, the higher the amount of La_2O_3 the higher the LAC until 2.506 MeV. This makes the suitability for S5, S4, S2, S1 and then S3 in descending order within this energy range. It can also be noticed that at energy between 2.506 to 4 MeV (Figure 2), density plays its role; the glass with higher density shows higher value of LAC. This makes S5 – S1 in a deceasing order of suitability, with S5 more suitable for radiation shielding in this region. For energies above 4 MeV until 15 Mev, S3 continues to lead the suitability series. For this reason, S3 is more suitable for radiation shielding at energies lower than 0.1 MeV and above 4 MeV while S5 is more suitable for energies from 0.1 to 4 MeV based on their LAC.

This result shows dependance of LAC on the compositions of a material and also on the energy of the incident radiation.

Mass Attenuation Coefficient (MAC): The higher the mass attenuation coefficient, the higher the protection ability of glass. The variation of MAC at various energies due to addition of La_2O_3 is shown in (Figure 3) and (Figure 4) for gamma ray photon energies within 0 -1 MeV and 1 – 15 MeV respectively.

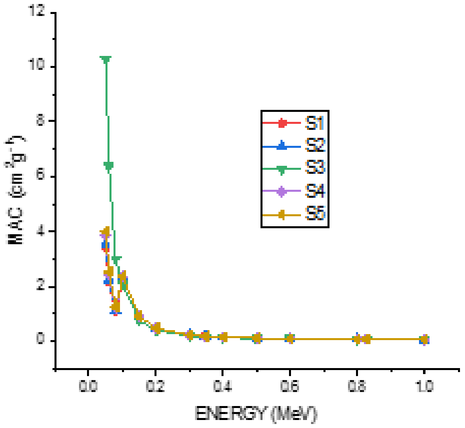

Fig 3: Variation of MAC with photon energies between 0 - 1 MeV

It can be seen from (Figure 3) that as the radiation incidents on the glasses, the mass attenuation coefficients of all the glass decrease drastically as the energy increases up to ~ 0.08 MeV (80 keV), the MAC then started to increase to reaches a local maxima of 2.357, 2.359, 2.085, 2.362 and 2.364 cm^2g^{-1} at energy ~ 0.1 MeV for S1, - S5 respectively. It then started to decrease at a lower rate until 1.0 MeV. It can also be noticed that at lower energies, S3, with La_2O_3 concentration of 1 mol% has the highest mass attenuation coefficient and undergone the gradual increase between 0.03 to 0.04 MeV. In the energy range between 1 – 15 MeV as shown in (Figure 4), the mass attenuation coefficients of all the glasses decreases with increasing energy and the rate becomes slower until the curves eventually flatten around 8 – 10 MeV and then gradually started to increase very slowly. However, The MAC of S3 is seen to be lowest at 1 MeV but highest for all the energy ranges above 4 MeV. These results show the dependence of MAC of a material on the radiation energy and also on the composition of the material, which is in agreement with other results published previously. It can also be noticed that increase in La_2O_3 results in increase in the MAC for all the samples within the energy ranges specified above.

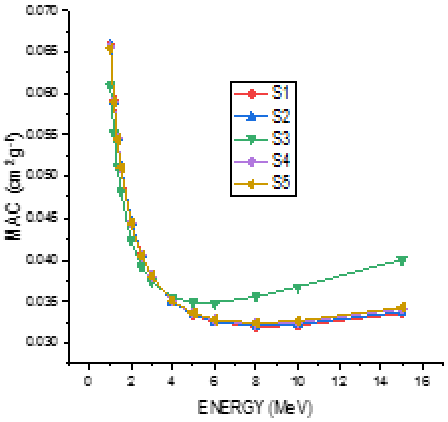

Fig 4: Variation of MAC with photon energies between 1 - 15 MeV

Half Value Layer, (HVL): Half Value Layer, (HVL), is one of the most important features that qualifies a material to be used as a radiation shielding. The lower the HVL of a material, the higher its ability to attenuate the incidence radiation (Al-Hadeethi & Sayyed, 2020) and vice versa. (Figure 5) and (Figure 6) show the HVL of S1 – S5 within 0 – 0.2 MeV and 0.2 – 15 MeV respectively.

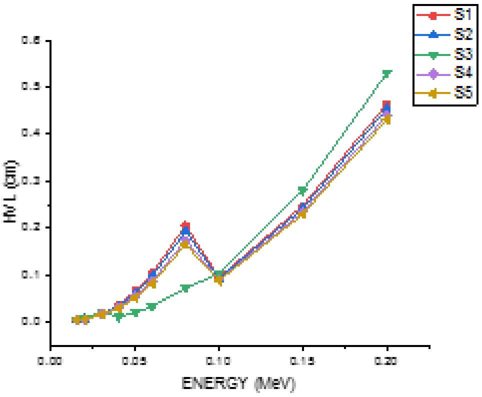

Fig 5: Variation of HVL with photon energies between 0 - 0.2 MeV

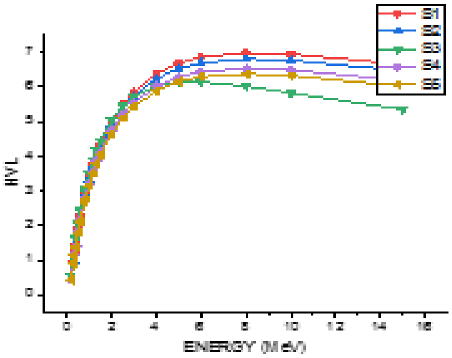

Fig 6: Variation of HVL with photon energies between 0.2 - 15 MeV.

It can be seen from (Figure 5) that, the higher the La_2O_3 the lower the HVL for all energies with exception of S3 which has lowest HVL within 0 – 0.1 MeV but highest HVL above 0.1 until 0.2 MeV. This indicates the betterment of the S3 at energies lower than 0.1 MeV (100 keV), but sample with higher La_2O_3 is more advantageous at energies above 100 keV. This result is true until when the energy exceeded 4 MeV, the S3 then has lowest HVL, and therefore more advantageous than other glasses. The minimum HVL values are 0.00479, 0.00465, 0.00388, 0.00443, 0.00431 cm for S1 - S5 at 0.015 MeV (15 keV) and the maximum values are 6.977, 6.799, 6.140, 6.528 and 6.372 cm for S1 – S5 which occurred at 8 MeV for S1, S2, S4 and S5, and at 6MeV for S3. At 0.347 MeV the lowest HVL values are 1.227, 1.201, 1.366, 1.164 and 1.141 cm for S1 – S5 respectively and the highest values are 2.283, 2.236, 2.432, 2.167 and 2.125 cm respectively. These values are lower than the HVL of ordinary concretes at this photon energy which is 3.877 cm and are lower than HVL of Ilmenite concrete, which is 2.651 cm (Al-Hadeethi & Sayyed, 2020).

Effective Atomic Number, Z_{eff}: Effective Atomic Number of a molecule plays the role that an atomic number plays in a single atom. The higher the effective atomic number of a material, the higher the number of interactions between the radiation and the atomic entities, and thus, the higher attenuation it offers to the radiation. The values of Effective Atomic Number of glasses S1 – S5 are shown in (Figure 7). It can be seen from (Figure 7) that the Z_{eff} of all the samples raises to a maximum of 73.311, 73.070, 59.641, 72.608 and 72.387 for S1 – S5 respectively between 0.015 – 0.02 MeV. It will then fall drastically with any slight increase in energy, between 0.02 - 0.08. This could be attributed to the photoelectric phenomenon, it will then rise up (as it can be seen in the portion shown by the arrow in the graph) until 0.1 MeV, then started to drop to its minimum at 1.5 MeV. From 1.5 - 15 MeV It will then start to increase gently up to 18.913, 19.179, 33.268, 19.703 and 19.961 for S1 – S5 respectively. This shows that the Z_{eff} is highest for all the glasses at energy lower than 0.02 MeV.

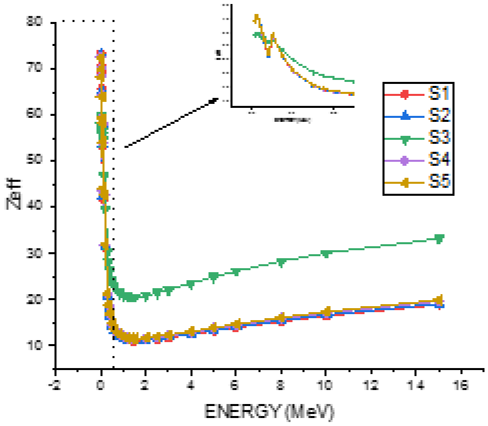

Fig 7: Variation of Z_{eff} with photon energies between 0 - 15 MeV

Conclusion: Glass samples S1, S2, S3, S4 and S5 with varying concentration of rare-earth metal Lanthanum Oxide, La_2O_3, are assessed for their radiation shielding features. The glasses have LAC, MAC and Z_{eff} higher than ordinary concrete and Ilmenite concrete, which are practically used for radiation shielding. This shows their applicability in photon attenuation, especially S3 which has the concentration of 1 mol% of La_2O_3, and has highest LAC, MAC and Z_{eff} and lowest HVL at energies below 0.1 MeV and above 4 MeV and S5 is more advantageous for energies within 0.1 - 4 MeV. We therefore concluded that, the addition of La_2O_3 into cobalt-doped borate glasses with chemical composition given above increases its radiation protection features and enables it to be better than some concretes.

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