

GAMMA-RAY INTERACTION STUDIES IN PHOSPHATE AND NON-PHOSPHATE GLASSES

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ABSTRACT

Phosphate and Non-phosphate glasses have been prepared by Melt quenching method. Gamma-ray mass attenuation coefficients for these glasses have been measured by using Narrow Beam Transmission Method with NaI(Tl) as detector. I_0 and d (g/cm^2) were measured and using exponential law of attenuation, the mass attenuation coefficients for each type of glass at discrete gamma energies i.e., 59.5 and 661.6 keV were calculated. The effective atomic numbers for photoelectric and coherent attenuation processes were also calculated for these glasses. Using mixture rule, μ/ρ values were calculated and were compared with the obtained experimental values. These both in turn were compared with theoretical values calculated using NIST XCom database. The theoretical values, calculated values and experimental results of the present work were found to be in good agreement.

Key words: Gamma-ray, Mass attenuation coefficients, Narrow beam transmission, Effective atomic number.

1. INTRODUCTION

In today's modern society with unimaginable, escalating global warming, the study of gamma ray interaction with matter is certainly the need of the hour. Sufficient importance is to be given to the applications in the area of radiation physics and dosimetry. This has led to in-depth

studies in wide variety of shields to protect against harmful radiation. As technology advances there is a need to develop materials which can be used under harsh conditions such as nuclear radiation exposure [1]. To choose an appropriate type of shielding material, the type of radiation that is being shielded, the energy of the radiation, and the level of attenuation have to be considered. Since Gamma rays are attenuated by processes which are functions of atomic number and mass of the material under consideration, materials which have high density and high atomic number such as Lead, Tungsten, Uranium etc., are generally sought after. Gamma radiation are best absorbed by atoms with heavy nuclei; the heavier the nucleus, the better the absorption [2]. The other prime factors which influence the choice of shielding materials are, cost of the material, weight of the material, and space available for the material. Lead shields are frequently used where space is limited or where only a small area of absorber is required [3]. But because of its high ductility, lead cannot be molded easily and does not retain a given shape

unless supported by a rigid material. Also owing to its low melting point 327.5°C , lead can be used only where the temperatures do not exceed its melting point. Compared to lead, Tungsten possesses far better shielding features. However, tungsten is many times more expensive than lead; therefore, it is preferred very rarely and is seldom used for massive shields. Hence a shift of research on alternative materials, which are cheap, yet durable and reliable, is seen in recent years. In situations where space is not a constraint and where structural strength is required, concrete is used. Most nuclear reactors use thick concrete shields to create a bioshield with a thin water cooled layer of lead on the inside to protect the porous concrete from the coolant inside [4].

But variations in composition and water contents, opacity to visible light makes concrete a less effective shield. One of the possible alternatives to concrete is glass material. Another advantage of choosing glass material is, the properties of glass compounds can be tailor made suitably by varying their composition and preparation techniques [5, 6]. Moreover they are comparatively less costly and hence to prove to be excellent shielding materials.

Gamma-ray shielding properties of various glasses have been studied theoretically and experimentally at different energies [7, 8, 9]. On the same lines studies on Bismuth-borate glasses [10], PbO-B₂O₃ glass system, Bi₂O₃-PbO-B₂O₃ glass systems [11], CaO-SrO-B₂O₃ glasses [12], PbO-BaO-B₂O₃ glass system [13], and PbO-SiO₂ glass system [14] have been carried out. All these reports showed that, compared to some standard

radiation shielding concretes, these glasses are far better option as shielding materials having an additional advantage of being light in weight, easy to handle and mold and also transparent to visible light. In light of the above studies, in the present work the authors tried to focus on mass attenuation coefficients of four new glass systems under two heads, phosphate and non-phosphate glasses, as alternative shielding materials, at two different gamma ray energies 59.5 and 661.6keV. The effective atomic number has proved to be convenient parameter for interpreting the attenuation of gamma ray by a compound of different elemental composition. Hence, effective atomic numbers for these glasses for photoelectric and coherent attenuation processes at these energies are also calculated.

1.1 REASONS FOR CHOOSING PHOSPHATE GLASS SYSTEM:

SiO₂, GeO₂, P₂O₅ and B₂O₃ are the four classic Zachariasen glass forming oxides. The glass former component in phosphate glasses is P₂O₅. Phosphate glasses are technologically and biologically important materials as they generally have a higher thermal expansion coefficient(α), lower melting temperature, bio-compatibility and a lower glass transition temperature 'T_g' than silicate and borate glasses [15,16]. However, phosphate glasses have poor chemical durability which often limits their practical applications. This component can be overcome by adding certain oxides to the phosphate glasses such as PbO [17], Al₂O₃ and Fe₂O₃ [18, 19]. In recent years, there has been lot of research on improving the physical properties and the chemical

durability of alkali phosphate glasses by adding different metal oxides of high valence cations like Al^{3+} , Ti^{4+} , Bi^{3+} , etc., because of the formation of relatively stable $M^{3+}-O-P$ bond [20]. These ions modify thermo-mechanical, physical and structural properties, basically due to change in glass structural network, through formation of cross-linked bonds.

Hence two Phosphate glass systems containing metal oxide, Al_2O_3 , alkaline metal oxide, K_2O and Alkaline earth metal oxide SrO/BaO was chosen for the present study.

1.2 REASONS FOR CHOOSING NON PHOSPHATE GLASS SYSTEM:

Contrary to the above two, a third system i.e., Tellurite glass system was studied. The primary structural units of tellurite glasses are TeO_4 trigonal bipyramidals and TeO_3 trigonal pyramidals where a change from TeO_4 to TeO_3 groups occurs with increasing content of modifying oxide [21]. An oxygen atom introduced into glasses with high TeO_2 content opens a $Te-O-Te$ bridge and changes two TeO_4 into TeO_3 units. Each of these TeO_3 units has two terminal $Te-O$ bonds with partial double bond character [22].

Tellurium dioxide is also a conditional glass former, which means it will form a glass with small molar% additions of a second compound such as an oxide or halide. TeO_2 glasses have high refractive indices and transmit into the mid-infrared part of the electromagnetic spectrum, therefore they are of technological interest for optical waveguides. Tellurite glasses have also been shown to

exhibit Raman gain up to 30 times that of silica, useful in optical fibre amplification [23]. Hence Lanthanoid oxide Er_2O_3 was introduced into Tellurite glass system along with Alkali metal oxide K_2CO_3 and transition metal oxide Nb_2O_5 .

A completely new combination containing alkali metal halogen LiF , Metalloid H_3BO_3 and Lanthanoid Oxide, Tb_2O_3 was chosen as the fourth system.

2. EXPERIMENTAL PROCEDURE

Analytical grade chemicals were used as starting materials. The usual way of obtaining a glass is quenching a melt obtained by fusion of one or more raw materials, which is known as the conventional melt quenching technique. High purity chemicals of $Al(PO_3)_3$, $Ba(PO_3)_2$, KH_2PO_4 , SrF_2 , AlF_3 , KF and Ln_2O_3 ($Ln = Nd$ and Er) were used as starting materials. After being well grounded in a mortar with a pestle, the mixed reagents were put into a platinum crucible and kept in an electric furnace around 1050 to 1100 $^{\circ}C$ for 1 hour and then poured onto a preheated brass plate. Then the formed glasses were annealed at 350 $^{\circ}C$ for ten hours to remove thermal stains. Finally, the glass samples were polished with silicon carbide abrasive paper and finally with a FIS polishing film to achieve very smooth surface.

Thickness measurement was carried out by a micrometer. The densities of the prepared glasses were measured by Archimedes principle using benzene as the immersion liquid. The chemical composition, label, density and thickness of all samples are given in table 1.

The total photon attenuation cross-section is measured in narrow beam geometry set up used by that of the Davison and Evans [24] as shown in the fig.1. In this procedure both source and detector are highly collimated, and this is achieved by using graded slits. A narrow beam of photons from two slits was made to fall on the sample material and transmitted beam was further collimated and collected by detector which was adequately shielded from external radiation. This was achieved using proper lead shielding around the source and the detector. Radioactive source ^{137}Cs (661.6 keV) of strength 10 mCi obtained from BARC Mumbai, India and ^{241}Am (59.5 keV) of strength 30mCi obtained from Amersham U.K. were used in the present investigation. The detector was a 2inch x 2inch NaI (Tl) crystal covered by aluminum foils of appropriate thickness and mounted on a RCA 8055 photomultiplier tube. The output pulses were amplified and fed through a 8K multichannel analyzer to a preset time scaler. The resolution and linearity of the spectrometer were studied using standard sources. The resolutions at full width at half maximum were 8% at 661.6 keV. Counts were noted with and without sample in the gamma ray beam and corrections were applied for background. The mass attenuation coefficient (μ/ρ) was then calculated. The measurement was repeated at least for ten times and determined the mean value of μ/ρ .

3. THE METHOD OF COMPUTATION

The elemental cross-section per electron is said to be directly proportional to Z^m where m depends on the type of process considered. A compound or a mixture may

be considered as a single element with an effective atomic number \bar{Z} given by,

$$\bar{Z} = \left[\sum_i \alpha_i Z_i^{m-1} \right]^{\frac{1}{m-1}} \quad (1)$$

where Z_i is the atomic number and α_i is the fractional content of i^{th} element in the compound or mixture. The mass attenuation coefficient (μ/ρ) can be expressed using the mixture rule [25-26]

$$\frac{\mu}{\rho}(E) = n_0 \left[K_{photo}(E) \bar{Z}^{m-1} + K_{coh}(E) \bar{Z}^{n-1} + {}_e\sigma_{KN} \right] \quad (2)$$

Where n_0 is the electron density, ${}_e\sigma_{KN}$ is Klein nishina cross-section and K is a constant at a given energy. The validity of these equations has been critically examined by Jackson and Hawkes [27] and it has been pointed out that they have no sound theoretical foundations. Nonetheless, there are many applications based on these equations over a considerable range of energy and atomic number Z [28].

The variation of elemental photon cross-section per atom (${}_a\sigma$) with atomic number was studied using the relationship

$${}_a\sigma = K(E) Z^m \quad (3)$$

The partial mass attenuation coefficient for a compound was assumed to be

$$\frac{\mu_{\tau}}{\rho} = K_{\text{photo}}(E)(\bar{Y})^p \quad (4)$$

with

$$\bar{Y} = n_0 \bar{Z}^{m-1}$$

A plot of $\ln\left(\frac{\mu_{\tau}}{\rho}\right)$ versus $\ln(\bar{Y})$ gives a straight line of slope p . This regression, using the calculated values of $\ln\left(\frac{\mu_{\tau}}{\rho}\right)$ and (\bar{Y}) for all the compounds was calculated for a number of values of m and the value corresponding to $p = 1$ was interpolated.

When $p = 1$, the equation resolved into the relationship given in equation (2). Similar calculations were made for attenuation coefficient due to coherent scattering. It has been pointed out [29-30] that equation (2) does not describe variation of the coherent, incoherent scattering and atomic photo effect to the required accuracy unless the exponents are allowed to be dependent on both E and Z . The total mass attenuation coefficient may be calculated from

$$\frac{\mu}{\rho}(E) = n_0 \left[e \sigma_{\text{KN}} + 103.3 \left(\frac{\bar{Z}^{m-1}}{E^{3.65}} \right) + 6.9 \left(\frac{\bar{Z}^{n-1}}{E^{2.01}} \right) \right] \quad (5)$$

4. Results and Discussion

The Z -exponents and effective atomic numbers for photoelectric process and

coherent process are given in table 2. Z -exponents and effective atomic numbers at the energy 661.6keV are greater than that at energy 59.5keV. This confirms the dependence of effective atomic number on the energy under consideration. Table 3 gives mass attenuation coefficients of all glass samples obtained from experimentation, calculated from equation 5 and theoretical X-Com values obtained from computer data base [31, 32]. The table also gives the values at two different energies under consideration, i.e., 59.5KeV and 661.6KeV.

An excellent correlation among experimental, calculated and theoretical values is observed. As expected the mass attenuation coefficient is more at low energy than at high energy. Moreover, there is considerable change in mass attenuation coefficient of the samples under consideration at low energy. Fig.2 shows the variation of mass attenuation coefficient (μ/ρ) with glass composition for energies 59.5 and 661.6 keV. From fig.2, mass attenuation coefficient is varying with glass composition with the following trend, NP1 > P2 > P1 > NP2 for 59.5keV energy. The sample NP1 consists of Te, Nb and Er elements and their K Jump energies are 31.81, 18.99 and 57.49keV respectively. The high experimental value of mass attenuation coefficient i.e., 4.591cm²/gm at 59.5keV energy for the sample NP1 may be attributed to the K Jump energy of Erbium which is a heavy rare earth element of Yttrium group. Adding to that all the other components of the NP₁ glass system are high Z elements when compared to the elements present in the other three glass systems. Moreover the

effective atomic number is also highest for this glass system as shown in table 2. Hence though the percentage of concentration of Erbium is less, this glass system has high value of mass attenuation coefficient when compared to the other three glass systems under consideration. Also addition of Erbium gives pink colour to the glass hence a coloured novel shielding glass could be prepared.

All the components and their percentages of samples P_1 and P_2 are the same except for the presence of comparatively high Z element Barium in P_2 in place of Strontium in P_1 . The K jump energy of Barium is 37.44 keV whereas K jump energy of strontium is only 16.1keV. These two factors may be responsible for greater mass attenuation coefficient of P_2 than that of P_1 .

Sample NP_2 consists of Terbium and its K jump energy is 52keV. Though the K jump energy is very much nearer to the energy source under consideration, the mass attenuation coefficient of NP_2 is less than that of P_1 . Also the mass attenuation coefficient of NP_2 is surprisingly less compared to other glass samples. This is because the percentage of the element Terbium is very less and also all the other components of this fourth glass system are low Z elements.

5. CONCLUSIONS

NP_1 glass sample having high mass attenuation coefficient at 59.5keV is the most promising radiation shield. At 59.5keV NP_2 is not suitable for radiation shielding. At 661.6 keV energy for all the studied glass samples there is no significant change in

mass attenuation coefficient. Hence glasses having considerable percentage of high Z elements consisting of lanthanoid oxides with high K jump value can be regarded as most suitable shielding materials at low energies.

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Figure 1. Schematic Diagram of Narrow Beam Geometric Setup.

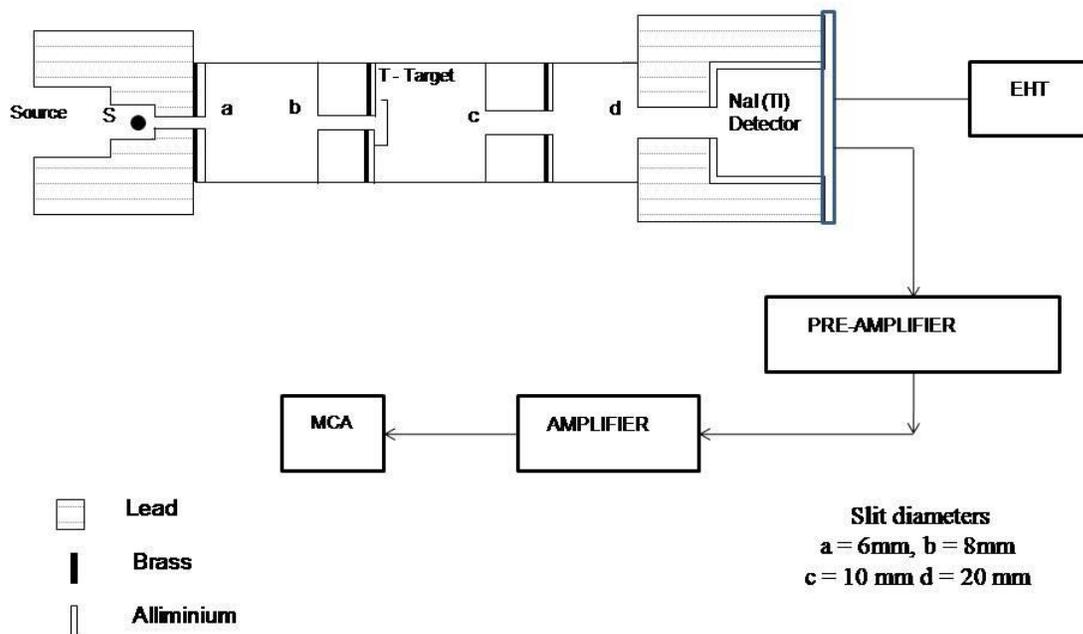


Figure 2. Variation of mass attenuation coefficient with glass composition for energies 59.5 and 661.6 keV.

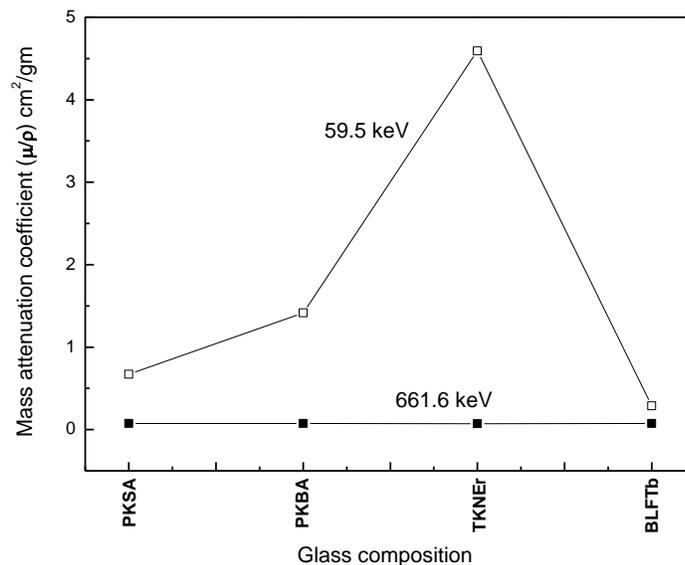


Table 1. Glass composition, label, density and thickness of the glass samples used in the present work

Glass	Exponents and Effective atomic numbers							
	59.5 keV				661.6 keV			
	Exponents		Z-	Z-	Exponents		Z-	Z-
	n	m	effective coherent	effective photo	n	m	effective coherent	effective photo
P ₁	2.41	4.5	16.32	22.06	2.62	4.98	16.89	23.21
P ₂	2.48	4.38	22.33	32.37	2.82	4.91	24.33	34.47
NP ₁	2.58	4.44	39.24	44.76	2.78	4.95	40.15	45.57
NP ₂	2.61	4.51	8.22	15.12	2.72	4.85	8.39	16.97

Table 2. Z-exponents, Z-effective coherent and Z-effective photo of the four glass systems

S.No	Chemical composition of glass	Label	Density gm/cc	Thickness mm
1	59 P ₂ O ₅ + 17 K ₂ O + 15 SrO + 9 Al ₂ O ₃	P ₁	2.674	4.82
2	59 P ₂ O ₅ + 17 K ₂ O + 15 BaO + 9 Al ₂ O ₃	P ₂	2.767	4.48
3	69TeO ₂ +15K ₂ CO ₃ +15Nb ₂ O ₅ +1Er ₂ O ₃	NP ₁	4.846	4.28
4	49.5H ₃ BO ₃ +49.5LiF+1Tb ₄ O ₇	NP ₂	2.748	3.73

Table 3. Experimental, Calculated and Theoretical Mass attenuation coefficients at energies 59.5 and 661.6keV

Glass	Mass attenuation coefficients (μ/ρ , cm ² /gm)					
	59.5 keV			661.6 keV		
	Experimental	Calculated	Theoretical X-Com	Experimental	Calculated	Theoretical X-com
P ₁	0.672	0.679	0.685	0.074	0.075	0.075
P ₂	1.415	1.433	1.436	0.076	0.0769	0.0764
NP ₁	4.591	4.666	4.623	0.075	0.075	0.075
NP ₂	0.289	0.293	0.290	0.074	0.075	0.075