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Comprehensive study on physical, elastic and shielding properties of lead zinc phosphate glasses

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ABSTRACT

A series of ternary phosphate glasses in the form of (PbO)x(ZnO)60-x(P_2O_5)40 where x = 0–60 mol%, have been successfully prepared by conventional melt-quenching technique. The physical and elastic properties of the glasses have been investigated using pulse echo technique. The longitudinal and shear velocity of the glasses were measured using the MBS8000 ultrasonic data acquisition system at 10 MHz frequency in room temperature. The density, ultrasonic velocity and elastic moduli are found to be composition dependent and the correlation between the elastic moduli with the atomic packing density is discussed in detailed. The shielding parameters, mass attenuation coefficients, half value layers and exposure buildup factor (EBF) values have been computed using WinXCom program with the use of GP fitting method, and variation of shielding parameters are discussed for the effect of PbO addition into the glasses and photon energy. An increase in the density of the glasses results in a change in crosslink density. The sound velocity and elastic properties increased with PbO content and increase in Poisson's ratio trend suggests that the rigidity of the glasses has decreased. Besides, the replacement of ZnO by PbO causes an increase in mass attenuation coefficient, while the half value layer and the exposure buildup factor were decreased and these glasses has been potentially used as shielding material.

1. Introduction

Recently, research attention and interest on phosphate glasses has been encouraged for solid state laser applications by their various unique properties like low melting temperature, good chemical durability, high dielectric constant, low crystallization ability, good transmission for infrared rays with a wide range of wavelengths, high thermal stability and low phonon energy [1–4]. However, the hygroscopic nature and poor chemical durability of the phosphate glasses limit their practical applications in various fields. To counter the problem, the addition of some metal oxides in phosphate glass network improves the chemical durability and spontaneous the emission probabilities of the glasses [5].

A number of research have been carried out to improve the physical properties and the chemical durability of phosphate glasses by adding different types of metal oxides into the glass structure. As example, lead oxide (PbO) is a heavy metal oxide. Pb^{2+} ions incorporation in the phosphate glasses by improved the chemical stability through the formation of P—O—Pb bonds and enhanced the moisture resistant of the glasses [6]. PbO can play the dual role, one as a modifier in PbO_6 structural units and the other as a glass former in PbO_4 structural units [7]. Also, the chemical stability of lead phosphate glasses has been further improved through the creation of non-bridging oxygens (NBOs) atoms by incorporating a third metal oxide to the glass host [8–10].

Besides, other metal oxides such as CaO, MnO, ZnO, Al_2O_3, Ga_2O_3, In_2O_3 and Sc_2O_3 were substitute to the PbO-P_2O_5 glass system and found to be good glass stabilizers [11–17]. Furthermore, the non-linear optical effects of such glasses resemble of other PbO-P_2O_5 glass system doped with rare earth ions [18–20]. On the other hand, the disadvantaged of the glasses such as low chemical durability, extraordinary hygroscopic and volatile nature of lead phosphate glasses disallowed them from replacing the commercial glasses in a wide range of technological applications [21]. As example, the addition of As_2O_3 to the PbO-P_2O_5 glass system has improved the optical properties such as optical transparency in the blue color region [22]. Furthermore, As_2O_3 act as strong network former in the glass matrix and they are expected to affect the infrared transmission of phosphate glasses since the frequencies of some of the fundamental modes of As_2O_3 vibration structural groups place in the region of vibration of phosphate structural groups [23]. Besides, it is also expected that As_2O_3 groups may form a single arsenic-phosphorus-oxygen frame work and may strengthen the glass structure. The addition of As_2O_3 to the PbO-P_2O_5 glass makes them suitable

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for the extensive distance optical transmission with the low loss and right candidate for fiber Raman amplification [24].

In recent decades, ZnO-based materials have drawn the significant interest of researchers due to specific properties such as non-toxic, non-hygroscopic nature, low cost, direct wide band gap, intrinsic emitting property and large exciton binding energy. Lead zinc phosphate (PbO-ZnO-P_2O_5) glasses show significant improvement in the chemical and physical durability while maintaining low glass transformation and softening points [25]. Although there have been many studies of PbO-ZnO-P_2O_5 glasses, there are limited studies on lead-zinc phosphate glasses in terms of glass structure and shielding properties. Shielding properties depends upon element composition of a material and shielding capacity increases with increase in atomic number of elements. In addition to shielding properties, the toxicity of materials handling in nuclear installation has been considered at top level nowadays. Therefore, the PbO-ZnO-P_2O_5 based glasses are found superior shielding glasses than other PbO based glasses. The fabrication and characterization of ternary PbO-ZnO-P_2O_5 glasses exhibit a potential useful for glass-to-metal sealing applications in windows of nuclear reactors, isotope handling facility and any other high radiation exposure area, where in drastic exposure reduction is required. The specific applications of PbO-ZnO-P_2O_5 glasses make challenge to explore the knowledge of their relationships due to structural and shielding properties. The prime goal of the present work is to study the physical, structural and shielding properties of the PbO-ZnO-P_2O_5 glasses as a potential candidate for shielding applications.

2. Methodology

2.1. Physical properties

The series of ternary lead zinc phosphate, (PbO)x(ZnO)60-x(P_2O_5)40 glasses were prepared by weighing and mixing a stoichiometric mixture of high purity lead oxide, PbO (98.0%), zinc oxide, ZnO (99.0%) and phosphorus pentoxide, P_2O_5 (98.5%) in a 50 ml alumina crucible. Then, 15 g batches of each glass samples were weighed using an electronic digital weighing machine with an accuracy of ±0.0001 g. During the melting process, the precaution step was taken to minimize the loss of P_2O_5 by having two separate heating stages. Firstly, the crucible was placed into the electric furnace with temperature of 450 °C for 1 h to evaporate any water vapor trapped in the mixture. Then, the sample was heated at 1000 °C for 2 h, so that a chemical reaction occurred rapidly. However, the exact melting temperature depended on the glass composition. Then, the crucible contain melt was shaken every 15 min to increase the bubble acceleration rate because gas bubbles from the oxide would be trapped in the viscous glass melt during the process of melting. The cylindrical shaped mold with 20 mm in height, 20 mm in external diameter and 10 mm internal diameter had to be clean from dust and impurities by polishing it using sandpaper (ranging coarse grains to fine grains) and then using acetone to clean and wash it before the melt was poured into the stainless steel split mold. The solid transparent glass sample was then transferred to another electrical furnace for annealing process at 400 °C for 1 h to prevent the glass sample from cracking (thermal shock phenomenon). After 1 h, the furnace was switched off and the glass sample was left inside before it was taken out after cold to room temperature. Both the surfaces of the sample were cut about 8–10 mm in thickness using a diamond cutter for the ultrasonic measurement. Samples were polished using fine grains sandpapers and a digital vernier caliper was used to check on the thickness of the glass samples.

The amorphous nature of the glass samples was studies by X'Pert PRO MPD diffractometer (PANalytical, Philips) and the measurements were carried out with Ni-filtered Cu-Kα radiation (λ = 0.1542 nm) radiation as the X-ray source at 40 kV and 40 mA. The densities of the glass samples were measured by the Archimedes' method at room temperature using acetone as the immersion liquid (density; 0.79 g cm^{-3} at room temperature). Acetone was chosen as immersion liquid because it has low surface tension. Due to this reason, it will discourage air bubbles from adhering to the sample during immersion. Furthermore, acetone will not react with or be absorbed by the glass samples. Samples in the air and the acetone were weighed using an electronic weighing machine with an accuracy of ±0.01 g. By using pulse-echo technique, the values of ultrasonic wave velocities were obtained at room temperature by measuring the elapsed time between the initiation and the receipt of the pulse appearing on the screen of MATEC Model MBS8000 ultrasonic digital signal processing system with 10 MHz resonating frequency. Burnt honey was used as bonding material between X-cut and Y-cut transducers and the glass sample for generating and detecting the longitudinal and shear ultrasonic waves [26].

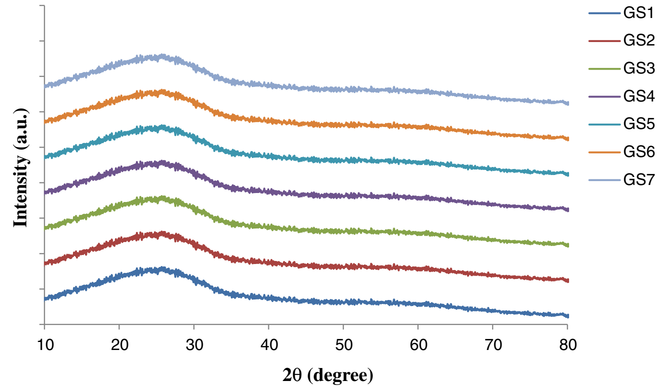

Fig. 1. XRD patterns of lead zinc phosphate glasses.

2.2. Shielding properties

2.2.1. Mass attenuation coefficient

The mass attenuation coefficient of the selected glass systems can be calculated using the mixture rule [27]

$$\mu/\rho = \sum_i w_i(\mu/\rho)_i \quad (1)$$

where w_i and $(\mu/\rho)_i$ were the fractional weight and the total mass attenuation coefficient of the ith constituent in the mixture respectively. The linear attenuation coefficient, μ (cm^{-1}) was evaluated by multiplying (μ/ρ) by the density of the glass. Mass attenuation coefficient (μ/ρ) of an element was determined by using WinXCom computer software [28,29]. This software provides attenuation coefficients for different interaction processes, such as coherent and incoherent scattering and pair production for several elements,

Table 1
Experimental values of density, longitudinal, shear ultrasonic wave velocity and elastic moduli of the lead zinc phosphate glasses.

Glass sample	PbO	ZnO	P_2O_5	Density, ρ (g cm^{-3}) ±0.001	Molar volume, V_m (cm^{-3}) ±0.04	Longitudinal, V_L (ms^{-1}) ±1	Shear, V_S (ms^{-1}) ±1
GS1	0	60	40	3.281	37.97	4376	2506
GS2	10	50	40	3.975	39.52	4171	2197
GS3	20	40	40	4.599	40.07	3852	2053
GS4	30	30	40	4.801	37.23	3658	1990
GS5	40	20	40	5.109	35.69	3259	1827
GS6	50	10	40	5.469	34.76	3043	1778
GS7	60	0	40	5.732	33.42	2873	1632