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Investigation of structural, thermal properties and shielding parameters for multicomponent borate glasses for gamma and neutron radiation shielding applications

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ABSTRACT

Multicomponent borate glasses with the chemical composition (60 − x) B_2O_3–10 Bi_2O_3–10 Al_2O_3–10 ZnO–10 Li_2O–(x) Dy_2O_3 or Tb_4O_7 (x = 0.5 mol%), and (60 − x − y) B_2O_3–10 Bi_2O_3–10 Al_2O_3–10 ZnO–10 Li_2O–(x) Dy_2O_3–(y) Tb_4O_7 (x = 0.25, 0.5, 0.75, 1.0, 1.5, and 2.0 mol%, y = 0.5 mol%) have been fabricated by a conventional melt-quenching technique and were characterized by X-ray diffraction (XRD), Attenuated Total reflectance-Fourier transform Infrared (ATR-FTIR) spectroscopy, Raman spectroscopy, thermo-gravimetric analysis (TGA), and differential scanning calorimetry (DSC). Also, the radiation shielding parameters such as mass attenuation coefficient (μ/ρ), half value layer (HVL), mean free path (MFP) and exposure buildup factor (EBF) values were explored within the energy range 0.015 MeV–15 MeV using both XCOM and MCNPX code to determine the penetration of gamma and neutron radiations in the prepared glasses. The main BO_3, BO_4, BiO_6, and ZnO_4 structural units and Al−O−Al bonds were confirmed by ATR-FTIR and Raman spectroscopy. Weight loss, and the glass transition (T_g), onset crystallization (T_x), and crystallization (T_c) temperatures were determined from TGA and DSC measurements, respectively. The stability of the glass against crystallization (ΔT) is varied within the temperature range 114–135 °C for the studied glasses. In addition, the shielding parameters like the (μ/ρ) values investigated using both MCNPX Monte Carlo and XCOM software are in good agreement with each other. The (μ/ρ) values calculated using XCOM software were used to evaluate the HVL and MFP in the photon energy range 0.015 MeV–15 MeV. It is found that all the synthesized glasses possess better shielding properties than ordinary concrete, zinc oxide soda lime silica glass and lead zinc phosphate glass indicating the high potentiality of the prepared glasses to be utilized as radiation shielding materials.

1. Introduction

Nowadays, due to the more extensive applications of radioactive isotopes in many fields such as nuclear power plants, radiotherapy, industries and agriculture there is an increasing interest among researchers in developing better radiation shielding materials to protect the humans, animals, and the environment against radiation exposure and dangerous effects of neutral radiations like gamma rays and neutrons [1–3]. Generally, radiation shielding involves incorporating a material between the radioactive source and the workers or the environment to reduce the exposure of the harmful radiations and as a consequence, the materials to be used for shielding must possess space homogeneity of density and composition. Conventionally, different types of concretes are currently in use as radiation shielding materials as they are inexpensive and easy to design in various shapes and sizes [4]. But, concretes show several disadvantages such as cracks formation with prolonged exposure to nuclear radiations, decrement in density and mechanical strength due to trapped water content and chemical damage etc. [5]. Further, concrete is not transparent to visible light and workers or onlookers cannot monitor the inside of such structures. Thus

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shielding materials such as glasses due to their high transparency in the visible region are certainly preferable over opaque concretes for absorbing radiations like gamma-rays and neutrons. Moreover, glasses physical and chemical properties can be easily altered by changing the composition and choosing different manufacturing methods. Glasses production cost is also not expensive and easily can be prepared in large sizes with high space homogeneity.

Though various lead (Pb)-based glasses are commercially available for nuclear radiation shielding applications; recently, a great deal of concern has been shown about toxic effects of Pb to human health, the well-being of living organisms and environment. So, numerous studies have been reported in the literature to replace Pb-based glasses with borate [6], borosilicate [7], tellurite [8], boro-tellurite [9] and borate-tellurite-silicate [10] glasses for radiation shielding applications. For this purpose, Barium (Ba) and Bismuth (Bi) are the two glass components that were evaluated recently [6–8,10], and in Ref. [11], photon interactions and shielding properties of silicate glasses containing Bi_2O_3, BaO and PbO were reported and it was found that the silicate glass containing Bi_2O_3 exhibits superior shielding properties followed by PbO and BaO glass systems, respectively, and all the studied glass systems were better than the standard shielding concretes. Due to their high atomic numbers, Bi and Ba elements enhance the gamma ray shielding properties of the glasses. Further, bismuth-based silicate and borate glass systems show better γ-radiation shielding properties in terms of their linear attenuation coefficient, effective atomic number and exposure build-up factor in comparison to lead-based glass systems [12].

The mass attenuation coefficient (μ/ρ), the effective atomic number (Z_{eff}), half value layer (HVL), the mean free path (MFP), the effective electron density ($N_{e,eff}$), and total interaction cross-section are the important parameters for evaluating the penetration of gamma-rays and neutrons in matter. Further, exact values of μ/ρ and $N_{e,eff}$ are required for the study of γ-radiation interactions with matter for medical and radiological applications. Here, Z_{eff} is the same as the atomic number, Z of the elements and provides information about how the radiation interacts with the absorbing medium. Also, for the desirable shielding glass, a higher value of an atomic number, total interaction cross-section and minimum irradiation effects on structural, mechanical and optical properties are required [1–12].

B_2O_3 is one of the commonly used glass formers with a lower price (cost-effectiveness) and can solely by itself form a glass at lower melting points with good optical transparency and show large glass forming tendency, good thermal stability, and moderate rare-earth (RE) ion solubility [13]. Boron (B) atom possesses a higher strength of the (B—O) bond including smaller cation size, and borate-rich glasses show coexistence of BO_4 tetrahedral units and BO_3 triangular units in their network structure. Bi_2O_3 is a well-known heavy metal oxide (HMO) and the glasses containing Bi_2O_3 possess low rates of crystallization, higher density, high refractive index, non-toxicity, high radioactive resistance, extended infrared optical transparency, large optical basicity, high optical susceptibility, high polarizability and moisture resistance [14,15]. Bi^{3+} ion has high Z_{eff} and due to small field strength of Bi^{3+} ion, though it cannot form glass itself, when added to glass formers like B_2O_3, Bi_2O_3 acts as a glass former at higher concentrations and serves as a network modifier at lower concentrations in a relatively wide compositional range [15,16]. In glasses, aluminum oxide (Al_2O_3) addition improves the mechanical strength, chemical durability and also reduces the glass thermal expansion coefficient [17]. Depending on its concentration, ZnO plays a dual role both as network former (at higher concentration) and glass modifier (at a lower concentration) and it imparts extended UV optical transparency, lower melting temperatures and enhanced glass forming region in oxide glasses [13,18]. It is well known that when introduced with modifiers, such as Li_2O or other alkali oxides, the covalent network of amorphous B_2O_3 changes considerably and rearranged through the transformation of some BO_3 structural units of the borate network to BO_4 units due to the formation of non-bridging oxygens [19]. Very recently, for white light emitting diodes (W-LEDs) application, RE ions like Dy^{3+}-doped or Dy^{3+}/Tb^{3+} co-doped multicomponent glasses attracted much attention among researchers due to an intense blue (470–500 nm) and yellow (570–600 nm) emission bands of Dy^{3+} ion, which arise from $^4F_{9/2} \rightarrow {}^6H_{15/2}$ and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ transitions, respectively, along with a weak red luminescence band (645–665 nm, $^4F_{9/2} \rightarrow {}^6H_{11/2}$ transition) [20–23]. Tb^{3+} ion shows an intense green emission at ~545 nm ($^5D_4 \rightarrow {}^7F_5$ transition) with an experimental branching ratio, $\beta_{exp.} > 50\%$ for green lasers and as a green light emitting source in W-LED applications [22–24].

In the present work, we have investigated structural and radiation shielding properties for several multicomponent borate glasses with singly Dy^{3+}, Tb^{3+}-doped, and codoped Tb^{3+}/Dy^{3+} ions along with the host glass. The glasses amorphous nature and network structure are examined by using XRD, and FT-IR, Raman spectroscopy techniques. Thermal properties of the glasses are evaluated from the thermogravimetric analysis (TGA), and differential scanning calorimetry (DSC) profiles. We also reported the radiation attenuation properties of the synthesized glasses by using MCNPX (version 2.4.0) Monte Carlo code to calculate the mass attenuation coefficient (μ/ρ) for A–I glasses. The validation of modeled MCNPX code has been obtained by comparing the results with standard XCOM data. The calculated mass attenuation coefficient values were then used to evaluate different γ-ray shielding parameters such as half value layer (HVL), mean free path (MFP), exposure buildup factors (EBF) for the prepared glasses.

2. Experimental

2.1. Glass synthesis and characterization

The conventional melt quenching technique was used to prepare the glasses with appropriate chemicals in 20 g batch each separately, at 950 °C for 40 min. The starting materials used were high purity B_2O_3 (99.98%), Bi_2O_3 (99.995%), Al_2O_3 (99.99%) ZnO (99.99%), Li_2CO_3 (99.99%), Dy_2O_3 (99.99%), and Tb_4O_7 (99.999%). All the chemicals were purchased from Sigma–Aldrich Company. The obtained glasses were transparent and of good optical quality, having a diameter of 3–4 cm and a thickness of ~ 0.5 cm. The glasses were annealed below the glass transition temperature, at 300 °C for 5 h in the air atmosphere to release the thermal stress and strain associated with these glasses during the quenching process and then allowed to cool slowly to ambient temperature. The nominal composition of the nine synthesized glasses is given in Table 1 and labeled as “A”, “B”, “C”, “D”, “E”, “F”, “G”, “H”, and “I”, respectively.

The density of the glasses was determined using the buoyancy method based on the Archimedes' principle with toluene as an immersion liquid to a precision of 0.001 g. For X-ray diffraction, structural and thermal analysis of the glasses, the types of equipment and parameters used were same as we described in our earlier publications [13,25]. In brief, the attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectra of the glass powders were measured over the 250–4000 cm^{-1} range by a Perkin Elmer Spectrum 100 FTIR spectrometer with a spectral resolution of ~4 cm^{-1}. The Raman spectra of the glasses were measured with a WITec alpha 300R Confocal Raman system equipped with cw doubled frequency Nd: YAG laser (532 nm, power 10 mW) as the excitation source. The Raman spectra were recorded within the spectral range of 0–3800 cm^{-1} for the Raman shift, with an integration time of 5 s for each particular Raman spectrum. The TGA and DSC measurements were performed with a Mettler Toledo TGA/DSC 1 HT Integrated Thermal Gravimetric Analyzer with high purity nitrogen as a carrier gas and a flow rate of 50 mL/min. For these measurements, about 20–30 mg glass powders were used in an alumina pan and the samples were heated from ambient temperature up to 1000 °C at a heating rate of 10 K/min., using Al_2O_3 as a reference sample. We performed the TGA/DSC (simultaneous) measurements

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