Journal of Non-Crystalline Solids 487 (2018) 53–59

Contents lists available at ScienceDirect

Journal of Non-Crystalline Solids

journal homepage: www.elsevier.com/locate/jnoncrysol

Structural, thermal, optical features and shielding parameters investigations of optical glasses for gamma radiation shielding and defense applications

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ARTICLE INFO

Keywords:
Optical glass
XRD
FTIR
Raman
TGA/DSC
Gamma-ray shielding

ABSTRACT

To design new radiation shielding materials instead of conventional materials such as concrete with improved performance in gamma-ray shielding in nuclear reactors, defense applications, and medical sciences, very recently, there exists an increasing interest among researchers. For this purpose, heavy metal oxides based optical glasses attracted much attention, nowadays. In this work, five glass samples with nominal composition of (70-x) B_2O_3-5SiO_2-5TeO_2-10ZnO-10Li_2O-xBaO (where x = 5, 10, 15, 20, and 25 mol%) were fabricated by melt quenching technique. The structural and thermal studies were carried out using XRD, SEM/EDAX, FTIR, Raman, and TGA/DSC measurements. Different shielding parameters such as mass attenuation coefficients, effective atomic number, mean free path and exposure buildup factor have been calculated in order to investigate the effectiveness of using the prepared glasses as radiation shielding materials. The half-value layer of the prepared glasses has been compared to different types of concretes. The results of shielding properties indicate that addition of BaO significantly enhances shielding capability of the prepared glasses against gamma radiation.

1. Introduction

Nowadays, tellurite glasses became well known as ideal materials for various scientific and technological applications because of their excellent chemical and physical characteristics like high refractive index, low melting point (800 °C), high dielectric constant, good chemical resistance and low photon energy [1–4]. It is recognized that TeO_2 is not a typical glass former; consequently, it requires the addition of other components to form a glass. The addition of WO_3 to tellurite glasses leads to an increase in the devitrification resistance and chemical stability, while the addition of PbO to tellurite glasses lowers the melting temperature. Furthermore, the addition of zinc oxide (ZnO) into tellurite glass network produces low rates of crystallization and increases the glass forming ability [5]. A considerable number of investigations have been reported on thermal, elastic, structural, electrical, shielding and optical properties of tellurite glasses [1,2,5–9]. Radiation shielding properties play a significant and major role in the selection of the glass for certain application because it supplies convenient details on the capability of a material to shield the radiation of specific energy. When dealt with concrete as a shielding material in nuclear reactors, it is found the most commonly used because of its distinguishable properties in that field. Economically it is cheap and adaptable to any building design. However, concrete has a number of disadvantages like non-transparency in the visible region, crack formation when exposed to radiation for long periods of time and change in its water content because of evaporation process at very high gamma-ray energies, leading to uncertainty in evaluations for protection designing. In consideration of this situation, it is compulsory to develop a superior gamma-ray radiation material, which can act effectively compare to concrete [10].

The glass is an amorphous solid that normally transparent, hard, brittle and pure to the elements of glass. Besides, glass also classified as a good material for the transmission of visible light. For that reason, glass can be a noble replacement for concrete as gamma-ray shielding material [11–13].

Interaction of radiation with shielding materials can be described by some essential parameters like the mass attenuation coefficient (μ/ρ), effective atomic number (Z_{eff}), half-value layer (HVL) and mean free path (MFP). The mass attenuation coefficient measures the rate of energy loss by gamma-ray as it penetrates a medium. It is an important tool utilized to obtain the effective atomic number and mean free path. On the other hand, the gamma-ray interaction with a medium can be described by the mean free path, which represents the average distance traveled by a photon in a medium before the interaction occurs, while

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https://doi.org/10.1016/j.jnoncrysol.2018.02.014
Received 7 December 2017; Received in revised form 28 January 2018; Accepted 15 February 2018

the half-value layer is the thickness of the material where one-half of the intensity of incident photon can be attenuated. It is worth to mention that the smaller values of MFP and HVL for glass samples show their higher shielding against gamma radiation [14,15]. The exposure buildup factor (EBF) is another important parameter in estimating the distribution of photon flux in an irradiated medium. It is defined as the photon buildup factor in which the quantity of interest is exposure and the detector response function is that of absorption in the air [9].

Though Pb-based glasses are useful for γ-ray radiation shielding applications because of their enhanced physical and chemical properties, their toxic nature is the main concern for the environment. Thus, very recently, there is an increasing interest in evaluating Pb-free glasses such as borate, silicate, tellurite, and zinc boro-tellurite glasses suitable for radiation shielding applications [16–25]. This work comes as continuity in the line of research work on this type of glasses. In this work, we used melt quenching technique to fabricate five glass samples with nominal composition (70-x)B_2O_3-5SiO_2-5TeO_2–10 ZnO-10Li_2O-xBaO (where x = 5, 10, 15, 20, and 25 mol%). To study the various structural units present in the glasses FTIR and Raman spectroscopy studies were performed. Thermal stability of the glasses was investigated by TGA and DSC measurements. Their shielding parameters (μ/ρ, Z_{eff}, HVL, and MFP) were calculated theoretically using XCOM program [25]. In addition, the EBF values are evaluated by the Geometric Progression (G-P) fitting method. The computational work of EBF and the G-P fitting parameters of the prepared glasses have been carried out by the well-known method of logarithmic interpolation from the equivalent atomic number (Z_{eq}). The computational work was done in three steps as follows. The first step deals with the calculation of the Z_{eq} values for the prepared glasses. The second step focuses on the computations of G-P fitting parameters while the values of EBF computations are accomplished in step three [26].

2. Experimental

2.1. Glass synthesis and characterization

In the present work, five glass samples with nominal composition (70-x)B_2O_3-5SiO_2-5TeO_2-10 ZnO-10Li_2O-xBaO (where x = 5, 10, 15, 20, and 25 mol%) were fabricated by melt quenching technique. For the glass synthesis, B_2O_3 (99.98%), SiO_2 (99.99%), TeO_2 (99.995%), ZnO (99.99%), Li_2CO_3 (99.99%), and $BaCO_3$ (≥99%) chemicals purchased from Sigma–Aldrich Company and Fisher Scientific were used as received. Here, as Li_2O and BaO precursors Li_2CO_3 and $BaCO_3$ have been selected with gravimetric factors of 2.473, and 1.287, respectively. In Table 1, the nominal chemical compositions and density values of the synthesized glasses are presented and labeled as "R1", "R2", "R3", "R4", and "R5", respectively, based on the BaO content from 5 to 25 mol%, for convenience. After thoroughly mixing the accurate amount of respective reagents for 15 g batch using an agate mortar and pestle, the materials were placed in a high purity alumina crucible and melted in an electrical furnace at 950–1090 °C depending on the glass composition for 45 min. The melt was then poured onto a stainless-steel plate and quickly pressed using another steel plate to obtain clear and transparent glasses with ~6 mm thickness. The obtained glasses were annealed for 5 h at 330 °C and then cool down to ambient temperature,

Table 1
Nominal compositions and density values of the synthesized glasses.

Glass code	B_2O_3	SiO_2	TeO_2	ZnO	Li_2O	BaO	Density (g/cm^3)
R1	65	5	5	10	10	5	2.953
R2	60	5	5	10	10	10	3.118
R3	55	5	5	10	10	15	3.275
R4	50	5	5	10	10	20	3.451
R5	45	5	5	10	10	25	3.602

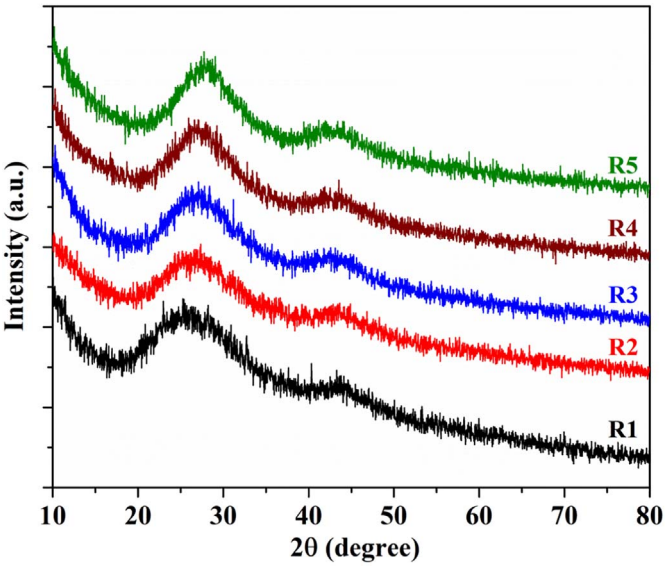

Fig. 1. XRD profiles for all the prepared glasses.

naturally. Few small pieces of each glass sample were crushed to acquire fine powder, which was then used for XRD, ATR-FTIR, Raman spectroscopy, and TGA/DSC measurements. For optical absorption measurements, the glass samples were cut and polished on both sides. Methods and equipment details used for the density (in solid form), XRD, SEM/EDAX, ATR-FTIR, Raman spectroscopy, and TGA/DSC measurements for the synthesized glass samples are the same as described in our recent publications [2,27]. The room temperature optical absorption spectra for all the glasses within the 200–2500 nm wavelength range were measured using a dual-beam spectrophotometer (Hitachi U-4100 UV–Vis-NIR) with spectral resolution 2 nm.

3. Results and discussion

Fig. 1 presents the XRD profiles for all the synthesized glasses. From this figure, amorphous nature of the glasses was confirmed with an absence of any crystalline peaks, except a few broad humps. The non-crystalline nature of the glass samples was also confirmed by SEM images, which showed uniform surface without any crystallites formation, and all elements present in the glass samples was confirmed by EDAX measurement (not shown here). Fig. 2 shows the FTIR spectra of R1–R5 glass samples within the wavenumber region 400 and 1600 cm^{-1}. Mainly, the vibrations of isolated BO_4 units and Li^+ or Ba^{2+} ions through glass network including stretching vibrations of ZnO_4^- units can be indicated by the IR bands at 400–580 cm^{-1} [28]. In pure borate glasses, the IR bands at 683–699 cm^{-1} could be due to bending mode vibrations of B–O–B linkages in various borate groups. In our studied borotellurite glasses, the identified 683–699 cm^{-1} bands are assigned to TeO_3 trigonal pyramids (tps) units [2]. The B—O bond stretching vibrations of tetrahedral BO_4^- units are identified with the presence of 883–897 cm^{-1} and 988–1058 cm^{-1} IR bands, respectively [29]. The IR bands at 1230–1246 cm^{-1} are assigned to B—O bond stretching vibrations and B—O bridging between B_3O_6 and BO_3 triangles and symmetric stretching vibration of BO_3^- units. The 1318–1328 cm^{-1} IR bands are ascribed to stretching vibrations of B—O bonds of the trigonal $(BO_3)^{3-}$ units in various types of borate groups [28,30].

The measured Raman spectra for all the prepared glasses are shown in Fig. 3. The low-frequency Raman bands at ~142 cm^{-1} can be usually assigned to boson frequency region, however, in our synthesized glasses, these are assigned to BO_3 and BO_4 vibrational modes due to alkali/alkaline borates. For R1 sample, the Raman band at 239 cm^{-1} indicates Metal–Oxygen (M–O) rotational and vibrational modes [28].