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The heavy metal oxide glasses within the WO_3-MoO_3-TeO_2 system to investigate the shielding properties of radiation applications

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ABSTRACT

A new series of heavy metal oxide (HMO) glasses in the $10WO_3$-$xMoO_3$-$(90\text{-}x)TeO_2$, x = 10–40 mol% system have been developed using melt quenching technique to investigate their potentiality for radiation shielding applications. Gamma attenuation parameters such as mass attenuation coefficient (MAC) and half value layer (HVL) were measured in a good transmission geometry. The MACs were then used to calculate effective atomic number (Z_{eff}). A HPGe detector was used to detect gamma rays of energies 81 keV, 276 keV, 303 keV, 356 keV and 384 keV emitted through a ^{133}Ba radioactive source. MACs, HVLs and Z_{eff}s were also calculated in a continuous energy region. One more relevant gamma shielding parameter, the buildup factor was also estimated for the glass samples in a continuous energy region at different penetration depths. Moreover, the developed glasses were investigated with respect to their shielding ability against fast neutron and charged particle radiation in the light of the calculated ranges for electron, proton and alpha particle interactions and macroscopic removal cross sections for fast neutrons. The results were compared with ordinary concrete and a lead (Pb) based glass sample wherever possible. It was concluded that the $10WO_3$-$10MoO_3$-$80TeO_2$ glass sample showed excellent shielding properties when compared to other glasses as well as concrete and Pb-based shielding glass.

1. Introduction

High energy ionizing radiations (indirectly ionizing e.g. photons, neutrons and directly ionizing e.g. electrons, protons, alpha particles) have found wide applications in radiation technologies, medical applications, nuclear physics and in space technology. Therefore, proper radiation shielding for experimenters and surrounding equipment has still become a problem. Accordingly, studies not only on the two types of most frequently encountered indirectly ionizing radiations i.e. photons and neutrons, but also on the directly ionizing radiations such as charged particles became the subject of interest due to required appropriate safeguards to evade from the radiation hazards (Shamshad et al., 2017).

Concrete containing different aggregates have widely been used in radiation shielding applications since they are widely available and have a low cost when compared to other shielding materials (Kurudirek, 2014). However, when exposed to high radiation levels the concretes get heated and therefore the water they contain is lost leading reduction in both density and structural strength of concrete (Issa and Mostafa, 2017). Furthermore, concrete is opaque to visible light making it impossible to observe what is going on behind. In this regard, glass with its double function of being transparent to visible light and absorbing gamma rays and neutrons provides an alternative radiation shield for observers and/or experimenters (Manohara et al., 2009). Glass also offers some certain advantages such as ease of modification of its properties by changing its composition and ability of vitrification for long-term storage of radioactive waste products.

Several researchers have contributed in the field to find out radiation shielding properties of various glass systems including lead (Pb) as a constituent for which serious environmental concerns still arise. Mostafa et al. have investigated gamma ray shielding properties of PbO-B_2O_3-P_2O_5 glasses doped with WO_3 and concluded that effective atomic number and electron density increase with the increasing concentration of WO_3 (Mostafa et al., 2017). Matori et al. have studied physical, elastic and shielding properties of lead zinc phosphate glasses and reached to a conclusion that replacement of ZnO by PbO has significantly increased the radiation attenuation capability of glass samples (Matori et al., 2017). Kumar measured mass attenuation coefficients of PbO-Li_2O-B_2O_3 glasses at different gamma energies and $Pb_3B_4O_9$ glass was found to be the most effective shield (Kumar, 2017).

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Table 1
Nominal compositions and density values of WO_3-MoO_3-TeO_2 glasses.

Sample ID	Composition	Density (g/cm^3)
W10M40T50	$10WO_3$-$40MoO_3$-$50TeO_2$	5.111
W10M30T60	$10WO_3$-$30MoO_3$-$60TeO_2$	5.255
W10M20T70	$10WO_3$-$20MoO_3$-$70TeO_2$	5.439
W10M10T80	$10WO_3$-$10MoO_3$-$80TeO_2$	5.627

However, Pb-based glasses have begun to lose their popularity because of their toxicity and very recently studies headed towards the synthesis of heavy metal oxide glasses, especially tellurite glasses, for radiation shielding applications due to their unique properties, such as non-toxicity, high density, high refractive index and high chemical resistance (Çelikbilek et al., 2013; Ersundu et al., 2012). Several studies showed that tellurite glasses provide successful radiation shielding efficiency and addition of WO_3 or B_2O_3 enhances this behavior. However, almost all of these studies were realized using WinXCom program with a theoretical approach and no experimental work has been carried out (Sayyed, 2016a,b, 2017; Sayyed et al., 2017). Therefore, in this study, which is a part of an ongoing research project on HMO-based glasses, it is aimed to investigate the radiation shielding properties of glasses within the WO_3-MoO_3-TeO_2 system both experimentally and theoretically by using a ^{133}Ba radioactive source and WinXCom program under different energies.

2. Theoretical procedure

The well-known Beer–Lambert law can be used to obtain the mass attenuation coefficient of the material under study:

$$I = I_0 e^{-\mu t} \tag{1}$$

$$\mu_m = \left(\frac{\mu}{\rho}\right) = \frac{\ln(I_0/I)}{\rho t} \tag{2}$$

where, I and I_0 are attenuated and unattenuated beam intensities, μ (cm^{-1}) and μ_m (cm^{2}g^{-1}) are linear and mass attenuation coefficients, respectively. t (cm) is the linear thickness and ρ (g.cm^{-3}) is the density of the material.

Half-value layer (HVL) is calculated using the linear attenuation coefficient (μ) through the following relation:

$$HVL = \frac{\ln(2)}{\mu} = \frac{0.693}{\mu} \tag{3}$$

The total atomic cross section (σ_t) for materials can be obtained from the measured mass attenuation coefficients μ_m using the following equation:

$$\sigma_a = \frac{\mu_m}{N_A \sum_i w_i/A_i}\left(\frac{b}{atom}\right) \tag{4}$$

here N_A is the Avogadro number, A_i is the atomic weight and w_i is the weight fraction of each element present in the material.

The atomic cross sections (σ_a) of materials can be further used to obtain the Z_{eff} using the below linear logarithmic interpolation equation:

$$Z_{eff} = \frac{Z_1(\log\sigma_2 - \log\sigma) + Z_2(\log\sigma - \log\sigma_1)}{\log\sigma_2 - \log\sigma_1} \tag{5}$$

where σ_1 and σ_2 are the total atomic cross sections of elements at the selected energies, in between σ of the material lies. Z_1 and Z_2 are atomic numbers of the elements corresponding to the σ_1 and σ_2, respectively.

WinXCom computer program was used to calculate the total mass attenuation coefficients (MACs) theoretically (Gerward et al., 2004).

Energy absorption and exposure buildup factors for the glass samples were calculated by the five parameter G–P fitting method for which detailed explanation is given elsewhere (Kurudirek and Topçuoğlu, 2011; Singh and Badiger, 2014; Karabul et al., 2015). The ranges for charged particles were calculated using SRIM Monte Carlo software (Ziegler et al., 2010). Fast neutron removal cross sections were calculated as described elsewhere (El-Khayatt, 2010).

3. Experimental

Glasses in the $10WO_3$-$x$$MoO_3$-$(90\text{-}x)TeO_2$ ($x = 10$–40 mol%) system were prepared by using a conventional melt quenching method starting from high purity chemicals of TeO_2 (99.99% purity, Alfa Aesar), WO_3 (99.8% purity, Alfa Aesar) and MoO_3 (99.95% purity, Alfa Aesar). After weighing and thoroughly mixing, batches of 5 g in weight were melted in a platinum crucible with a closed lid at 800 °C for 15 min. Vitreous samples were obtained by pouring the melt into pre-heated stainless steel molds. Then the samples were annealed near the glass transition temperature for 2 h to remove internal stresses. Finally the glasses were cut, ground and polished to obtain samples with plan-parallel surfaces having an average thickness of 1.5 mm for further radiation shielding measurements. The amorphous nature of as-cast samples was verified by X-ray diffraction (XRD) analysis with powdered samples by a Bruker™D8 Advanced Series powder diffractometer using Cu K_α radiation in the 2θ range from 10 to 90. Density values (ρ) of the synthesized glasses was measured at room temperature by the Archimedes' principle using distilled water as an immersion liquid and a digital balance of sensitivity 10^{-4} g. Density values were obtained by three repeated measurements with an error of ± 0.1%. Nominal compositions and densities of WO_3-MoO_3-TeO_2 glasses are listed in Table 1.

A 10 mCi ^{133}Ba radioactive point source was used to obtain gamma rays of 80.8, 276.4, 302.8, 356 and 383.8 keV. A HPGe detector (an active area of 200 mm^2, Be window with a thickness of 0.127 mm and a radius of 13.97 mm, dead layer thickness 500 Å, Ge crystal with a thickness of 7 mm and a radius of 7.981 mm, detector bias voltage −1500 V) was used to detect gamma rays reaching out detector before and after attenuating in the material. Detector has a resolution of

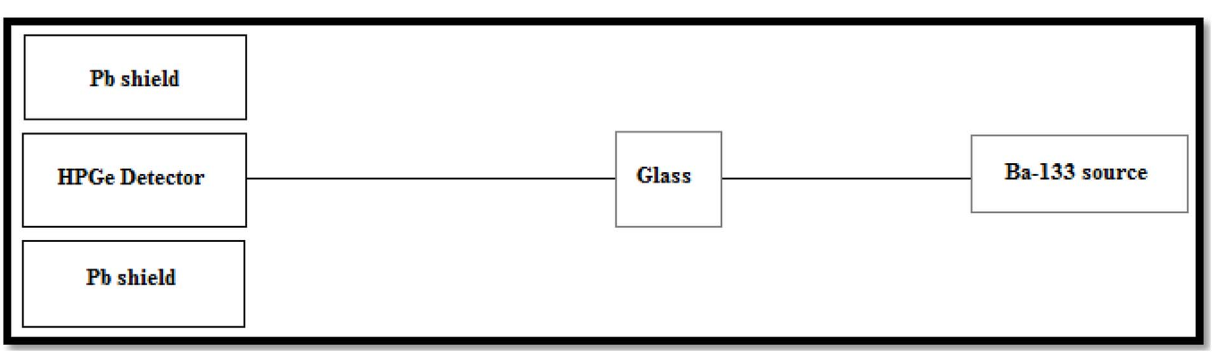

Fig. 1. Transmission geometry used in experiments.