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Influence of manganese doping on elastic and structural properties of silica borotellurite glass

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ABSTRACT

The quaternary glass system tellurium dioxide, boron trioxide, silicon dioxide and manganese dioxide where x = 0.00 to 0.05 M fraction has been successfully prepared via conventional melt and quench-casting technique. The density was determined by Archimedes method with distilled water as buoyant liquid. The results showed that the density decreases and molar volume increases with respect to manganese concentration. FTIR spectral studies suggest that glass network is mainly built up of boron trioxide, tetrahedral boron, tellurium trioxide, trioxotellurate and silicon dioxide structural units. The ultrasonic velocities and elastic moduli were recorded using a pulse-echo method at frequency 5 MHz at room temperature. Some other physical properties such as Debye temperature, softening temperature, microhardness, Poisson's ratio, fugacity, fractal bond connectivity and Cauchy relation were determined. Results indicate that these parameters depend upon the composition of the glass system and the dopant concentration, MnO_2 inside the glass system.

1. Introduction

Elastic properties have shown the stress-strain relationship in the material. According to William and Scott, correlated qualitatively the elastic moduli of oxide glasses with chemical composition [1]. Elastic moduli of materials are related to separation distance atoms, inversely proportional to the fourth power of atomic spacing and strength of the material [2]. According to Makishima and Mackenzie, elastic moduli of material are a function as average strength of the chemical bonds and packing density in the glass system [3]. More than that, when the elastic moduli of the materials increases, the strength of the material also increases [4]. The elastic moduli of glasses system are directly related to inter-atomic forces, potentials, and packing density of their oxide constituents give a short and mid-range structure [5–7].

Tellurite glasses are interesting due to their various unique properties such as low melting points, hygroscopic properties, high refractive index, wideband infrared transmittance and good glass stability from other oxide glass formers [8–11]. Borate glass can be used as addition in this glass network because of their unique behaviour such as utilized in low alkaline borosilicate glasses, good matrix material, low temperature sealing glasses [12–14]. Borate glass is also suitable to be used as non-linear optical component in fiber optics application. Transition metals have been widely used for magnetic, electric properties, mechanical, and optical due to their possible application in this various field. Manganese, which is made up of paramagnetic, is one of the transition metals that is suitable to be applied as a modifier because it can confer properties for example electrical, optical, thermal and magnetic and can change glass structure.

In this paper, $\{[(TeO_2)_{0.7}(B_2O_3)_{0.3}]_{0.8}[SiO_2]_{0.2}\}_{1-x}\{MnO_2\}_x$ glass system at different molar fraction, x = 0, 0.01, 0.02, 0.03, 0.04, and 0.05 was investigated and characterized by means of ultrasonic velocities, FTIR, density, and molar volume, in order to obtain detailed physical, structural information and elastic properties about glass network and the effect of manganese content role in this glass system. Since there is no study had been done on silica borotellurite glass doped with manganese oxide, the essential and useful characterization as well as measurement of this glass system can unveils crucial information for possible application of this glass system in the future.

2. Experimental

A series of silica borotellurite glass in the form $\{[(TeO_2)_{0.7}(B_2O_3)_{0.3}]_{0.8}[SiO_2]_{0.2}\}_{1-x}\{MnO_2\}_x$ with x value had been varied from 0 to 0.05 M fraction have been successfully prepared via melt quenching technique. The purity of TeO_2 (Alfa Aesar, 99.99%), B_2O_3 (Alfa Aesar, 98.5%), MnO_2 (Alfa Aesar, 99.99%) and SiO_2 from rice husk ash (98.85%). All the chemicals were weighed by using an electronic balance. The silica used was extracted from the rice husk. Rice husk were washed with normal water and distilled water to rid dirt and impurities. Cleaned rice husk were stirred with 2.0 M of

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hydrochloric acid (HCl) for 20 min and heated at 110 °C for 3 h. The rice husk was then rinsed with distilled water to remove the HCl acid. Then the rice husk was dried in oven at 110 °C for 3 h. Finally, the rice husk was burned at 700 °C for 6 h to obtain white rice husk ash. Then, the mixture was mixed together using mortar and pestle. The mixture was put in alumina crucible and preheated at 400 °C for 1 h. After that, the mixture undergoes melting process for 3 h at temperature 1100 °C. Next, the melted molten was poured into a cylindrical shape stainless steel mould that had been preheated. The sample was transferred back into the furnace for annealing process at 400 °C for 2 h. The sample was left in the furnace for overnight to be cooled at room temperature. The glass sample was cut into 5 mm thickness using diamond cutter before being polished using sand paper to get parallel surface for both sides. The density of the glass was determined by Archimedes method with distilled water as buoyant liquid by using Electronic Densimeter MD-3005. This accuracy of this instrument is ± 0.0001 g. The Fourier Transform Infrared (FTIR) spectra were obtained by using Perkin Elmer FTIR Spectrophotometer in the range of 280–4000 cm^{-1}. A spectral resolution of FTIR spectroscopy was ± 1 cm^{-1}. Both longitudinal and shear ultrasonic velocities were measured by using Ritec Ram-5000 Snap Ultrasonic System at room temperature by a pulse-echo method. The ultrasonic velocities can be calculated using $v = 2d/\triangle t$ where d is thickness of the glass sample and $\triangle t$ is the time taken between two consecutive echoes. The uncertainty is estimated to be about ± 1%. All the glasses samples were measured for five times for accuracy and were taken at a frequency of 5 MHz.

3. Results and discussion

3.1. Density and molar volume

The structural changes of the glass system can be determined by density and molar volume. The values of density (ρ) measured for the samples using Archimedes principle with probable error of ± 0.001 g/cm^3. The values of density of the prepared glass are found to be decreased from 4180 to 3710 kgm^{-3} with the increment of molar fraction of MnO_2 from 0 to 0.05 as shown in Fig. 1. The decrement in density of the prepared glass occurs are due to the substitution of high molecular weight TeO_2 (159.60 g/mol) with the low molecular weight MnO_2 (86.93 g/mol) molecules. As a lighter element is replaced with a heavy element, it will cause the decrement in a density, as density is directly proportional to a mass. As seen in the glass samples composition, TeO_2 is the highest percent that been used is the glass composition followed by B_2O_3 and SiO_2. Thus, as amount of TeO_2 and SiO_2 decreases while amount of MnO_2 increases, the overall density of the glass decreases. Besides that, the addition of MnO_2 into the glass system causes the

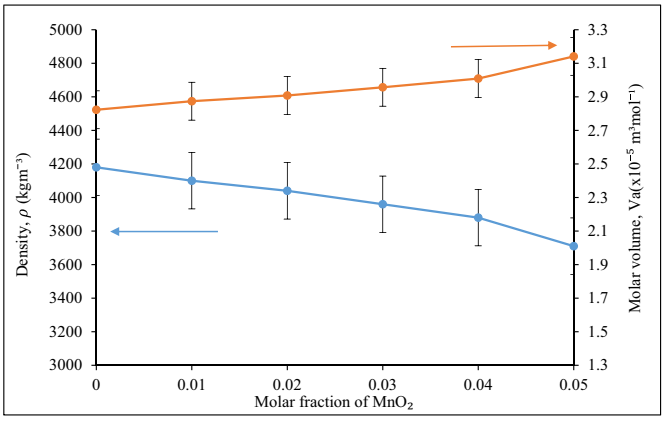

Fig. 1. Density and molar volume of $\{[(TeO_2)_{0.7}(B_2O_3)_{0.3}]_{0.8}[SiO_2]_{0.2}\}_{1-x}\{MnO_2\}_x$ glasses (lines are drawn as guides to the eyes).

structural changes which is changes in the coordination of boron and tellurite glass network. This changes with allows the creation of more non-bridging oxygen (NBO) from BO_4 to BO_3 and TeO_4 to TeO_3 that can be seen in FTIR result [15,16]. The molar volume, V_a of the glass sample is determined by using the following equation:

$$V_a = \frac{M}{\rho} \quad (1)$$

where M is the total molecular weight of the sample and ρ is density of the glass. The molar volume results as depicted in Table 2 shows that as the MnO_2 concentration increases, the value of molar volume is observed to increase from 2.823×10^{-5} to 3.141×10^{-5} $m^3 mol^{-1}$. This is also shown in Fig. 1. The increment of the molar volume is possibly attributed to the growth of free volume in the glass network, which leads to the change in the atomic mass and atomic volume of tellurite and manganese [17]. The atomic mass of tellurite and manganese are 127.60 and 54.94 respectively. According to previous research [16], the ionic radii of Mn^{3+} and Te^{3+} are 0.58 Å and 0.52 Å respectively. The same research also reported that manganese ion seems to exist in the Mn^{3+} with the coordination number of 5 while the presence of Te^{3+} state with coordination number equal to 3 has been confirmed by FTIR spectrum. Based on the results shown in Fig. 1, it can be inferred that the replacement of tellurite atoms with manganese atoms causes the gradual decrease of density with the respect to the increase of dopant concentration, as opposed to the change behaviour of the molar volume. The density and molar volume show opposite behaviour since molar volume and density theoretically inversed to each other.

3.2. Fourier transform Infrared (FTIR) spectroscopy

FTIR spectroscopy measurement was carried out to obtain and explain crucial behaviour and arrangement of the structural units in the studied glass. The FTIR spectra for glass sample are shown in Fig. 2. From the graph, it is observed that there are five obvious absorption bands in the spectra which are located in the range of 600–3150 cm^{-1}. The uncertainty for of this measurement is ± 10 cm^{-1}. The recorded FTIR spectra are characterized by superimposed broad peaks, which are deconvoluted using Origin 6.0 software. One of the typical deconvoluted FTIR spectra for the sample with x = 0.01 is shown in Fig. 3. Table 3 shows the deconvolution parameter, band centre and relative area for each peak of every glasses samples.

The peak observed at around 600–650 cm^{-1} in IR spectra is assigned to TeO_4 trigonal bipyramids [18]. The absorption peak in the region of 671–715 cm^{-1} is probably attributed to the presence of Te—O vibrations in trigonal pyramids in TeO_3 groups accompanied with non-bridging oxygen (NBO) [19,20]. Another IR peak is observed between 725 and 780 cm^{-1}, which originates from the B-O-B linkages in borate network [21]. The other region between 801 and 834 cm^{-1} is due to the B—O bond stretching of BO_4 units [21]. The well distinguished peak observed between 1071 and 1125 cm^{-1} in IR spectra is due to Si—O(Si) vibrations [22]. Bands at about 1232–1382 cm^{-1} can be attributed to a vibration of asymmetric stretching relaxation of the B—O bond of trigonal BO_3 units [21]. In the high wavenumber range of 3098–3119 cm^{-1} the band corresponds to hydrogen bond [23]. This band can only be seen for the sample with at x = 0 and 0.01 M fraction. It is worth to note that Mn vibration cannot be detected in the spectra may be due to limitations of the instrument or breaking of the bonds, indicating that the Mn acts as modifier in the glass system. Based on the IR spectra, the elastic properties of the glass system, can be related to the structural changes. Based on the Fig. 2, the FTIR spectra with the different molar fraction of manganese was nearly similar. The bands at 625 cm^{-1} were shifted towards higher wave number that signify an increasing TeO_4 number. Furthermore, bands arounds 680 cm^{-1} that were slightly shifted towards lower wave number imply the decreasing of TeO_3 unit with addition of MnO_2. It was likely because of the transformation of TeO_4 into TeO_3 unit. Absorption band near 806 cm^{-1}