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EFFECT OF SILVER ON THE PHYSICAL AND STRUCTURAL PROPERTIES OF LEAD NEODYMIUM BOROTELLURITE GLASS SYSTEM

(Kesan Perak atas Sifat Fizikal dan Struktur Sistem Kaca Plumbum Neodimium Borotelurit)

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Abstract

Neodymium doped borotellurite glass is well known to be one of the most promising candidate due to it various applications. Nd^{3+} doped lead borotellurite glasses containing silver were successfully been prepared by conventional melt-quenching method with the chemical composition (69.5-*x*)TeO_2-20B_2O_3-10PbO-0.5Nd_2O_3-*x*$AgNO_3$ (where *x* = 0, 0.5, 1.0, 1.5, 2.0 and 2.5 mol %). The physical properties such as density, molar volume and oxygen packing density were measured. Their structural properties of the glass system were studied via X-Ray Diffraction (XRD) analysis and Fourier Transform Infrared (FTIR) spectroscopy. From the result, the amorphous nature of the prepared glass samples have been confirmed through X-ray diffraction spectral analysis. The presence of Te-O-Te or O-Te-O, Ag-O, Te-O-Pb and B-O and characteristic of the hydrogen bond in the prepared glasses were explored through the FTIR spectral studies recorded in the 400-4000 cm^{-1} wave number range at room temperature. Meanwhile, the results of physical properties are found to vary with respect to concentration of $AgNO_3$ content. Some other results will be reported and discussed.

Keywords: neodymium, borotellurite, silver, physical properties, structural properties

Abstrak

Kaca borotelurit di dop neodimium merupakan salah satu bahan yang diyakini disebabkan oleh kepelbagaian aplikasinya. Kaca borotelurit di dop Nd^{3+} mengandungi perak telah berjaya disediakan dengan kaedah konvensional sepuh lindap dengan komposisi kimia (69.5-*x*)TeO_2-20B_2O_3-10PbO-0.5Nd_2O_3-*x*$AgNO_3$ (di mana x = 0, 0.5, 1.0, 1.5, 2.0 dan 2.5 mol%). Sifat-sifat fizikal seperti ketumpatan, isipadu molar dan ketumpatan padat oksigen telah diukur. Sifat-sifat struktur sistem kaca ini telah dikaji melalui analisis pembelauan sinar-X (XRD) dan spektroskopi inframerah transformasi Fourier (FTIR). Dari hasil kajian, sifat semulajadi amorfus sampel kaca yang disediakan telah disahkan melalui analisis spektra pembelauan sinar-X. Kehadiran Te-O-Te atau O-Te-O, Ag-O, Te-O-Pb, B-O dan ciri-ciri ikatan hidrogen dalam kaca yang disediakan telah diterokai melalui kajian spektra FTIR yang direkodkan dalam julat gelombang 400-4000 cm^{-1} pada suhu bilik. Sementara itu, hasil kajian bagi sifat fizikal didapati berubah mengikut kepekatan kandungan $AgNO_3$. Beberapa keputusan lain akan dilaporkan dan dibincangkan.

Kata kunci: neodimium, borotelurit, perak, sifat fizikal, sifat struktur

Introduction

The physical and structural studies of glass oxide incorporated with various rare earth (RE) draw much attention due to their important role in the development of many optical devices such as infrared laser, colour display devices, optical detectors, light converters and sensors [1]. Among the oxide glass, lead borotellurite glass systems were much attractive due to their high density, high refractive index [2], low phonon energies (~700 cm^{-1}), extended infrared transmittance (0.35-6.0 μm), large corrosion resistance, high mechanical strength and relatively high thermal stability [3, 4]. The glass hosts are excellent candidate for laser applications due to their low cost as well as ease to fabricate in various geometries [5]. In the literature there are relatively few reports on the structural properties of glass doped with Nd^{3+} ions [6, 7]. Recently study reported on the effects produced by the codoping with silver nitrate of glasses doped with various RE ions on the structural properties [8] that confer potential applications for these material. In this work, we prepared some Nd^{3+} doped lead borotellurite glass containing silver. The sample have a fixed Nd^{3+} and PbO content whereas the TeO_2:$AgNO_3$ content are varied. The physical properties by mean of their density, molar volume as well as the oxygen packing density (OPD) will be determined. Meanwhile, their structural study will be identify using the X-ray diffraction (XRD) and Fourier transform infrared (FTIR) spectroscopy. However, the aim of this study was to investigate the influence of silver on the physical and structural properties of the investigated materials.

Materials and Methods

Glasses with chemical composition (69.5-*x*)TeO_2-20B_2O_3-10PbO-0.5Nd_2O_3-*x*$AgNO_3$ (*x* = 0.0, 0.5, 1.0, 1.5, 2.0 and 2.5 mol%) were prepared by a conventional melt-quenching method using reagents of analytical grade. The chemicals of the required compositions of approximately 10g batches were weighed in an electrical balance and mixed thoroughly in milling process about 1 hour in order to obtain homogeneous mixtures. The mixture was melted at 1000 °C for 30 minutes in an electric box furnace. The mixture was then poured into stainless steel plate and immediately transferred into a preheated furnace for annealing process at temperature 400 °C for 3 hours before allowed to cool down gradually to room temperature. The glass samples of each composition were prepared and polished to obtain a planar faces for structural analysis. The composition of the investigated glasses are listed in Table 1.

Table 1. Chemical Composition of (69.5-*x*)TeO_2-20B_2O_3-10PbO-0.5Nd_2O_3-*x*$AgNO_3$ glass system

Sample No	Composition (mol%)				
	TeO_2	B_2O_3	PbO	Nd_2O_3	$AgNO_3$
S1	69.5	20	10	0.5	0.0
S2	69.0	20	10	0.5	0.5
S3	68.5	20	10	0.5	1.0
S4	68.0	20	10	0.5	1.5
S5	67.5	20	10	0.5	2.0
S6	67.0	20	10	0.5	2.5

The density of the glass samples was measured using distilled water as an immersion liquid by Archimedes' principle on a Electronic Densimeter MD-300S balance. Formula for calculating density, molar volume and oxygen packing density is given in Table 2. The structural properties of the glass samples were analyzed using X-Ray Diffraction (XRD) analysis which is performed at room temperature to confirmed its amorphous nature. The Fourier Transform Infrared (FTIR) spectroscopy absorption spectra of the glass samples were recorded on Perkin-Elmer Spectrum 100 FT-IR Spectrometer at room temperature in the wavenumber range 400-4000 cm^{-1} with a resolution of 4 cm^{-1}.

Table 2. Formulas for calculating physical properties of silver addition of lead borotellurite glass.

Eq. No.	Parameter	Formula	Description
1	Density (ρ)	$\rho = \left(\frac{W_a}{W_a - W_{dw}} \right) \rho_{dw}$	W_a - Weight of sample in air W_{dw} - Weight of sample in distilled water ρ_{dw} - Density of distilled water
2	Molar volume (V_m)	$V_m = \frac{M_{ave}}{\rho}$	M_{ave} - Average molecular weight ρ - Density
3	Oxygen packing density (OPD)	$1000n \times \frac{\rho}{M_{ave}}$	n - Number of oxygen atoms per formula ρ - Density M_{ave} - Average molecular weight

Results and Discussion

Physical properties

Table 3 shows the result of density and molar volume of Nd^{3+} doped lead borotellurite glass containing silver and Figure 1 shows the variation of density and molar volume with respect to silver content. As tabulated in the Table 3, the density of glass was found to increase from 4.969 gcm^{-3} to 5.053 gcm^{-3} with the increasing concentration of $AgNO_3$ from 0.0 mol% to 1.0 mol% before it start to decrease to 5.028 gcm^{-3} at 1.5 mol% of $AgNO_3$. By increasing of some amount of $AgNO_3$, the formation of non-bridging oxygen (NBO) take place and caused the modification of glass network brought extra compactness with higher density but shrinkage in free volume [9]. However, the density of glass seem to decreases slightly at the 1.0 mol% of $AgNO_3$. This may be due to the fact that an occupation of Ag between 1.0 to 1.5 mol% in the free space region in the network structure cause the compactness reduction of the glass network [10]. Meanwhile, the result of molar volume of the glass sample decrease from 29.642 cm^3/mol to 29.169 cm^3/mol with the increasing concentration of $AgNO_3$ from 0.0 mol% to 1.0 mol% before it start to increase to 29.324 cm^3/mol at 1.5 mol%. This trend of results are expected due to incorporation of Ag of radii 1.65 pm to replace Te of radii 1.23 pm tend to alter the glass network. This trend results has been observed by Yusof et al. [11].

Table 3. Physical properties of silver addition of lead borotellurite glass.

Physical Parameters	Unit	Glass Name					
		S1	S2	S3	S4	S5	S6
Density (ρ)	(g/cm^3)	4.969	4.980	5.053	5.028	5.031	5.071
Average molecular weight (M_{ave})	(g)	147.289	147.340	147.392	147.443	147.494	147.546
Molar volume (V_m)	(cm^3/mol)	29.642	29.586	29.169	29.324	29.317	29.096
Oxygen packing density (OPD)	(g.atom/L)	71.015	71.317	72.508	72.295	72.483	73.206

Meanwhile, the oxygen packing density (OPD) is the important parameter to explain the compactness of glass structure [12] and to measure of the tightness of the oxide network [13]. OPD measurements of all glass samples can be calculated using equation 3 from Table 2 and the results are listed in Table 3. As depicted in Figure 2, the OPD seen to vary with respect to silver content. From Figure 2 the OPD are found to increase from 71.015 g.atom/L to 72.508 g.atom/L with the increasing of $AgNO_3$ content from 0.0 mol% to 1.0 mol%. However, the values of oxygen packing density drop slightly to 72.295 g.atom/L as the $AgNO_3$ increased to 1.5 mol%. The results are then start to increase again from 72.483 g.atom/L to 73.206 g.atom/L as the $AgNO_3$ increase from 1.5 mol% to 2.5 mol%. This results show the similar trends to the density with respect to silver content [14].

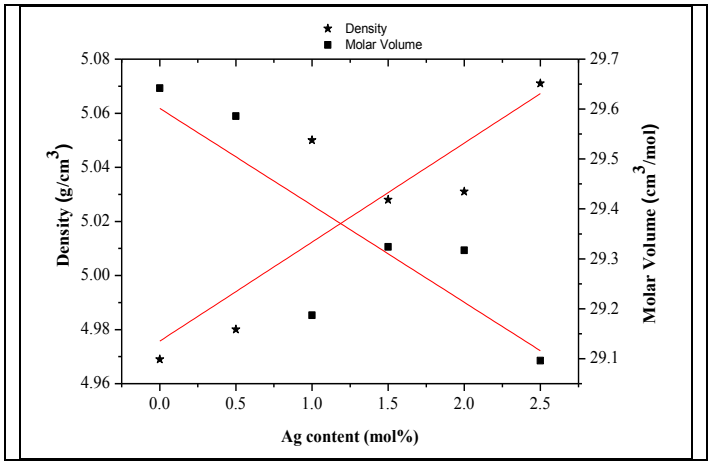

Figure 1. Variation of density and molar volume of silver addition of lead borotellurite glass

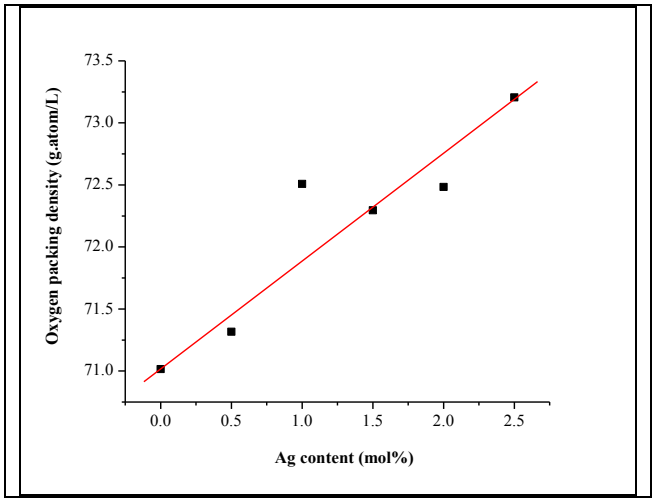

Figure 2. Variation of OPD of silver addition of lead borotellurite glass

Structural properties

The amorphous nature of the glass system (69.5-x)TeO_2-20B_2O_3-10PbO-0.5Nd_2O_3-$x$$AgNO_3$ with x varying from 0.0 to 2.5 mol% of $AgNO_3$ were confirmed through the x-ray diffraction pattern recorded in the range $10° \leq \theta \leq 90°$ as shown in Figure 3. As in Figure 3, there are no sharp peaks but broad hump diffusion been observed. This confirm the amorphous nature of the glass samples [15]. It can also be observed that the intensity of pattern in all glass samples is slightly different, thus indicating that the glass samples have a different degree of amorphousity. This result shows a good agreement with previous study [16].

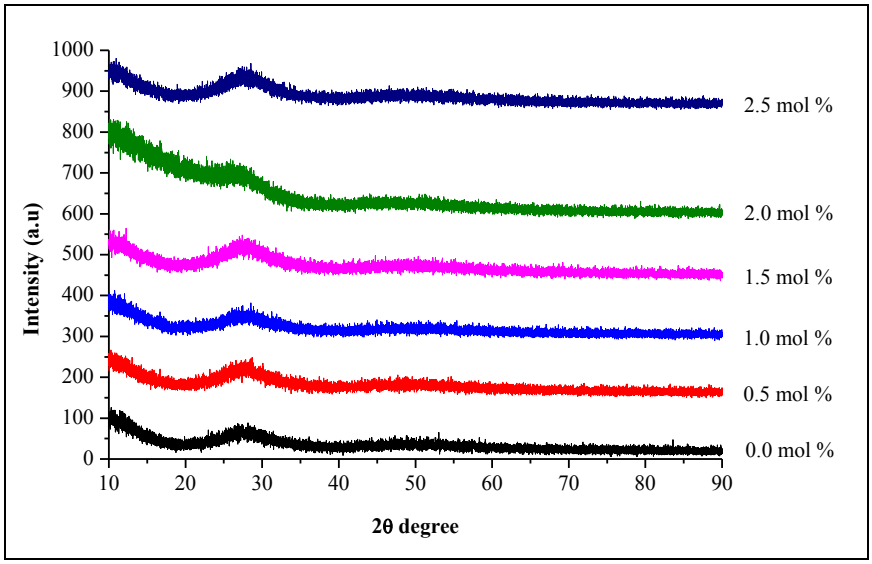

Figure 3. XRD pattern of silver addition of lead borotellurite glass.

In this work, FTIR spectroscopy was carried out in order to identify the existing functional groups peak in the glass networks. Therefore, Figure 4 shows the FTIR spectra of lead borotellurite glass in the range between 400-4000 cm^{-1} for different glass compositions. From Figure 4, the FTIR spectra of the borotellurite glasses show six significant IR absorptions bands respectively. Their peak assignments of these glasses are listed in Table 4 based on the literature data on the wavenumber ranges that related to the corresponding vibrations of the structural units in various glassy system.

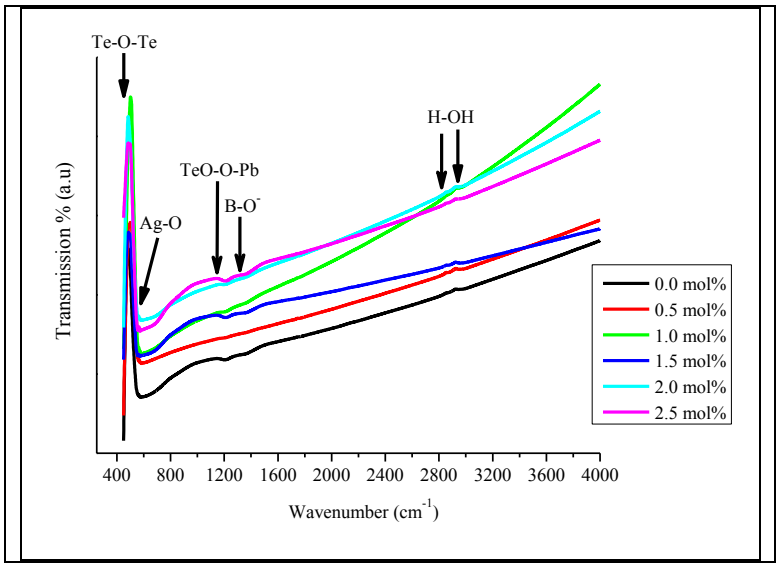

Figure 4. IR spectra of silver addition of lead borotellurite glass

As can be seen from Table 4, the bands are found in the regions around 460-476 cm^{-1}, 552-575 cm^{-1}, 1177-1209 cm^{-1}, 1350-1371 cm^{-1}, 2872-2880 cm^{-1} and 2940-2960 cm^{-1} wavelength respectively. The spectra bands observed between 460 cm^{-1} and 476 cm^{-1} are belongs to the Te-O-Te or O-Te-O symmetric stretching vibrations mode at corner sharing sites. This indicates that the vibrations of Te-O-Te or O-Te-O linkage in these glasses are corresponding to the transformation of TeO_4 group into TeO_3 groups [17]. Meanwhile, the bands around 552-575 cm^{-1} are assigned to symmetrical bending vibrations of Ag-O bond [18]. The IR absorption at around 1177-1209 cm^{-1} are attributed to the Te-O-Pb streching vibrations. This result indicates that the addition of PbO into the glass network could cause a change to the Te coordination polyhedron from TeO_4 trigonal bipyramid (tbp) to TeO_3 trigonalpyramid (tp) thus creating more non-bridging oxygen's (NBOs) [19]. The bands occurring in the spectral range 1350-1371 cm^{-1} are attributed to the B-O stretching vibration in BO_3 units. These bands are similar reported by Pawar et al. [20]. Finally, the absorption bands located around 2872-2880 cm^{-1} and 2940-2960 cm^{-1} are due to the characteristic of the hydrogen bond in the glass samples [21].

Table 4. IR band assignments of silver addition of lead borotellurite glass.

Characteristic Bands (cm^{-1})	**Assignment**
460 - 476	Stretching vibrations modes of Te-O-Te or O-Te-O
552 - 575	Bending vibrations of Ag-O
1177 - 1209	Streching vibrations of Te-O-Pb
1350 - 1371	B-O stretching vibration in BO_3
2872 - 2880	Hydrogen bond
2940 - 2960	Hydrogen bond

Conclusion

A series of glass of the system of $(69.5\text{-}x)TeO_2\text{-}20B_2O_3\text{-}10PbO\text{-}0.5Nd_2O_3\text{-}xAgNO_3$ where x = 0.0, 0.5, 1.0, 1.5, 2.0 and 2.5 mol% have been well fabricated as they are in a good quality as visualized. The density of the glasses increased from 4.969 gcm^{-3} to 5.053 gcm^{-3} while their molar volume values decreases from 29.642 cm^3/mol to 29.169 cm^3/mol with respect to $AgNO_3$ content. The oxygen packing density (OPD) show the same trend to the density as they vary from 71.015 g.atom/L to 73.206 g.atom/L with respect to $AgNO_3$ content. The XRD results confirmed fact the glasses obtained are amorphous in nature. The FTIR spectra indicated the presence of Te-O-Te or O-Te-O, Ag-O, Te-O-Pb and B-O functional group of the glass network.

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