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Ultrasonic velocities, elastic modulus and hardness of ternary Sb-V_2O_5-TeO_2 glasses

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ABSTRACT

Quantitative analysis was done to achieve more insight on the mechanical and structural properties of xSb-(60-x) V_2O_5-40TeO_2 ($0 \le x \le 15$ in mol%) glasses, prepared by melt quenching. Both room temperature longitudinal and shear ultrasonic velocities were measured by using pulse-echo method at frequency of 5 MHz. Using the ultrasonic velocities, longitudinal, shear, Young's and bulk elastic moduli, Poisson's ratio and Debye temperature were calculated and interpreted upon the number of bonds per unit volume n_b, oxygen ion packing density OPD and mean atomic volume $\overline{V}$. Results show anomalies with addition of Sb and show a behavior change at x = 12, confirming the changes in the rigidity and the content of non-bridging oxygens (NBOs). The obtained elastic moduli and Poisson's ratio confirm the previously reported trends of Makishima-Makenzie's theory with a relatively large deviations in amounts (up to 27%). These glasses were studied with respect to their Vicker's microhardness. The microhardness increases with Sb content and shows its maximum at x = 12 like elastic properties. A behavior change at x = 12 mol% in all recent data and also in previously reported optical band gap, oxygen molar volume and thermal stability implies to a drastic structural change with increasing Sb, associated to NBOs. Briefly, coincidence between the mechanical, structural, optical, and thermal data, suggests the glass with x = 12 as promising material in optical applications such as optical fibers because of its resistance against thermal and mechanical shocks.

1. Introduction

Generally, suggestion of a material for specific applications is a direct result of its chemical stability, ambient reliability and cost. These points, and also the compositional dependence of the material properties have led to numerous research works in the different subjects [1–6]. So, it should be mentioned that the study of optical, electrical, thermal, mechanical and structural properties are necessary, worthy and comprehensive to obtain more comprehension about the nature of materials (i.e. each study needs relevant and suitable tools and techniques). On the other hand, it should be mentioned that the glassy samples have rare properties (such as hardness, optical transparency, structural and mechanical strength at ambient temperature), which are not found in other materials. Interest in glasses has exclusively increased in recent years because of numerous advantages in electronic, nuclear, solar energy and acousto-optic devices [7–9].

Due to the ambient limitation of borate and phosphate glasses [7–10], tellurium-based oxide glasses are technologically interested because of:

(a) Their high chemical stability and their resistance to moisture for long periods [11].
(b) High glass forming ability with other transition metal oxides and even with alkaline elements [7].
(c) Suitable optical transmittance and other optical aspects [8].
(d) Low melting points [9].
(e) Their spreading applications as acousto-optical material [12], laser [12], photochromic glasses [13–14] and so forth.
(f) They could be prepared in different shapes and sizes and can accept different ions without inducing any crystallization.

Furthermore, the compositional dependence of acoustical properties is very informative in describing the glasses and giving some information about their microstructure and their dynamics. The structural and elastic properties are related to microscopic properties through the behavior of the network and the modifiers. Several works have been performed to measure and discuss the acoustical properties of different glass systems up on the structural changes [15,16]. Bahatti and Singh [17] have discussed the compositional dependence of ultrasonic velocities. The ultrasonic nondestructive pulse-echo technique has been found to be one of the informative techniques for mechanical characterization and also to estimate microstructures and phase changes as

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well as to evaluate elastic constants, microhardness, Poisson's ratio, ultrasonic attenuation coefficient and Debye temperature [18–21].

Therefore, one can characterize the different materials such as semiconducting glasses, glass ceramics, and bio-glasses by this non-destructive method. The propagation of longitudinal and transvers ultrasonic waves in the understudied glasses gives important and valuable information. Propagation of the acoustic waves in bulk glasses has attracted considerable interest to understand the mechanical properties [22]. The elastic moduli of glasses are affected by many physical parameters, which may somewhat in turn be investigated by ultrasonic measurements. The velocity of ultrasonic waves and hence the elastic moduli are particularly suitable for compositional characterization of glassy materials [23], which gains much important outputs.

As mentioned, elastic properties are very informative about the microstructure of the material and they are strongly and directly related to interatomic potentials [24–27]. It should be mentioned that, there are low T_g glasses with relatively high elastic moduli and vice versa [28,29].

The aim of a part of this article is to provide extensive study on the elastic properties of the studied glasses, in order to put more light on the intimate relationship existing between the elastic properties and the atomic network organization. Also, the structural deformation of glassy samples can be investigated by using the indentation analysis named as Vicker's microhardness test [30], which has important characteristics as its independency of the sample size, easy performance and localized plastic deformation around the indentation point; this test can complete the acoustical measurements. Elastic studies of some tellurovanadate glass systems such as the (50-x)V_2O_5-xBi_2O_3-50TeO_2 [31] and $(TeO_2)_{50}$-$(V_2O_5)_{50\text{-}x}$-$(TiO_2)_x$ [32] tellurovanadates been reported, but there is no report on the acoustical properties of the understudied samples.

To the best of our knowledge for the present samples (TeO_2-V_2O_5-Sb), some papers have been published on the optical [25], calorimetric [33,34], structural, and Makishima-Mackenzie's findings on the elastic properties of the same glassy system [35], which will be used in this work; but there are no any reports on the ultrasonic studies and Vicker's microhardness. In this article, we attempt first to deduce changes in the structure and elastic properties of this glass system, related to the substitution of V_2O_5 by Sb. Many compositional parameters such as the mean atomic volume, oxygen molar volume, the number of network bonds per unit volume, oxygen ion packing density, molar volume, Poisson's ratio, Debye temperature and the number of vibrating atoms per unit volume will be evaluated. In addition, the correlation between acoustic and compositional parameters will be discussed to gain insight into the structure of these glasses. Finally, the agreement between the evaluated values of theoretically calculated and experimentally measured elastic moduli is investigated. Elastic moduli, microhardness and other structural properties will be investigated to verify the validity of the Makishima-Mackenzie's theory for these glasses, with the idea of finding the samples owing higher potentials in applications.

In the light of these findings, the present study was undertaken with the aim of investigating the effect of Sb on the mechanical properties and the network structure of xSb-40TeO_2-(60-x)V_2O_5 glasses.

2. Experimental details

2.1. Sample preparation, XRD, and DSC

Ternary tellurium-based xSb-40TeO_2-(60-x)V_2O_5 glasses with x = 0, 5, 8, 10, 12, 15 mol% (nominated as TVSx) were prepared using rapid melt quenching method; it is worthy to mentioned that the obtained bulk glasses have been immediately annealed at a fixed temperature for 2 h to eliminate the thermal stresses (and so, reaching to more homogeneity of samples) and guarantee the reliability of the outputs of elastic experiments. More details of glass preparation process, structural characterization, titration, XRD, differential scanning calorimetry (DSC), glass density and some physical properties were precisely described in our previous published papers [25,33,34].

2.2. Vicker's microhardness

Bulk samples were highly polished at the surfaces and were used for Vicker's micro-hardness measurement using a Bareiss-Prufgeratebau Gm bH V-Test VTP 6045 BJ.07 Germany; load F of 0.1 HV (0.98 N) with a loading time of 10 s was employed. All of the under studied samples were tested with the same load and loading time, to ensure the crack-free indents. The indentation imprints were examined by optical microscopy owing a micrometer ruler. All measurements were done at room temperature. Four indentations in different areas of each sample were conducted in order to increase the accuracy of measurements. The hardness was calculated by the following equation:

$$H^{exp} = 0.1891\frac{F}{{d_{12}}^2} \tag{1}$$

where H^{exp} is Vicker's microhardness in Gpa, F is the applied load (force) in N, and d_{12} denotes the average value of the measured diagonals of the indentation imprint in m.

2.3. Ultrasonic aspects

In ultrasonic experiments, one needs a short time about ~15 min to measure the transverse and longitudinal wave velocities, and then the elastic properties of glasses can be readily measured. Furthermore, these ultrasonic echography (USE) outputs can be used to calculate elastic moduli and Poisson's ratio, in order to probe the structural changes occurring as a paramount interest for glass scientists.

The carefully surface-polished glasses were tested by the high precise pulse-echo technique to measure both longitudinal and shear ultrasonic velocities with the precise of ± 5 m/s. The accuracy in the reading of time intervals between two successive echoes (Δt) was ± 0.002%. These measurements was done using an ultrasonic pulse receiver (Panametrics model 5072PR) up on ASTM E494-10 standard at the frequency of 5 MHz. It should be mentioned that the ultrasonic method is of non-destructive testing techniques based on the propagation of ultrasonic waves inside the glass.

3. Results

3.1. Structural characterization by XRD

As reported in our previous works [25,33,34], the amorphous structure of the under studied glasses has been investigated and confirmed by XRD. Also, these reports imply on the justification of their amorphous nature by DSC analysis (existence of the baseline before glass transition [33–35].

3.2. Elastic velocities and moduli, Poisson's ratio and Debye temperature

Elastic modulus of a material is defined as the ratio of imposed stress on material to the strain produced by the material. As known, the pulse-echo technique is suitable and powerful technique to evaluate of elastic properties. The incident longitudinal and transvers waves can be transmitted in a material of thickness d and reflected back and forth at the two parallel surfaces, producing echo-signals. The first two echo signals corresponding to times t_1 and t_2, is regarded to evaluate the ultrasonic velocity U as:

$$U = \frac{2d}{t_2 - t_1} = \frac{2d}{\Delta t} \tag{2}$$

where Δt is the time interval between two successive echoes. Using the data of both measured longitudinal (U_l) and shear (U_S) velocities and also the glass density (ρ), Young's (E), bulk (K), shear (G) and longitudinal (L) moduli and also Poisson's ratio (σ) are achieved using