Journal of Non-Crystalline Solids xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Journal of Non-Crystalline Solids

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

Zero stress-optic bismuth oxide-based glass

E.C. Power[a], A.L. Paterson[a], U. Werner-Zwanziger[a], L.D. Ellis[a], J.W. Zwanziger[a,b,*]

[a] Department of Chemistry, Dalhousie University, Halifax, NS B3H 4R2, Canada
[b] Clean Technologies Research Institute, Dalhousie University, Halifax, NS B3H 4R2, Canada

ARTICLE INFO

Keywords:
Glass
Bismuth oxide
Zero stress-optic response
^{11}B NMR
Bismuth borate

ABSTRACT

In order to test the prediction that Bi_2O_3 could function as a replacement for PbO in preparing glass with zero stress-optic response, a series of bismuth boroaluminosilicate glasses of the form $Bi_2O_{3x}(0.86B_2O_3\text{-}0.10SiO_2\text{-}0.04Al_2O_3)_{1-x}$ were prepared. These glasses were then subjected to structural characterization and optical analysis. ^{11}B and ^{27}Al magic angle spinning nuclear magnetic resonance spectroscopy, Raman spectroscopy and optical absorption spectroscopy were used to track the structural evolution of the glasses across the series, while the Sénarmont method was employed to determine the stress-optic coefficient. Experiments indicated a zero stress-optic composition containing roughly 59 mol% Bi_2O_3. Various model glass structures were evaluated in the context of our previous model involving bond lengths and coordination numbers, and it was found that the prediction of this model was reasonable but that the elasto-optic response of the Bi-O bonds appears to be weak compared to that in Pb-O bonds.

1. Introduction

Similarities between the structural and chemical behaviour of Bi^{3+} in glass and Pb^{2+}, such as the high polarizability of both heavy metals [1], suggest Bi_2O_3 as an environmentally benign replacement for PbO [2–4]. Like lead oxide-based glasses, bismuth oxide glasses melt at characteristically low temperatures and have high densities. Additionally, Bi_2O_3-based glasses have attractive optical properties including high refractive indices, IR transmissivity and strong non-linear optical response [1,2,5-9]. In contrast to lead oxide, however, they typically have absorption band edges and hence narrower transparency windows.

One area where bismuth has been been little explored as a replacement for lead is in the preparation of zero stress-optic glass. The stress-optic response of glass refers to the birefringence induced when mechanical stress is applied, and can be positive, negative, or zero. Most oxide glasses have zero stress-optic response with about 50 mol % PbO content, meaning that the change in index of refraction in the stress direction is equal to the change perpendicular to the stress direction, at this composition. The d/N_C model we proposed predicts this composition, as well as a variety of lead-free alternatives [10]. In this model, cation-anion bond-length d is a proxy for bond metallicity, leading to more polarizable and less directional bonding [11,12], and N_C is the cation coordination number. Empirically, we found that the molar averaged d/N_C value, that is,

$$\sum_i x_i \left(\frac{d}{N_C}\right) \tag{1}$$

where x_i is the mole fraction of cation i, is equal to about 0.5 Å at the zero stress-optic composition, less for positive response, and more for negative response. The validity of this approach has been demonstrated by several studies [10,12-17]. Our original work predicted that Bi_2O_3 could be an alternative to PbO to prepare zero stress-optic glass [10], and the purpose of the present contribution is to investigate this prediction in detail.

Bismuth oxide can be a more difficult additive to use, as compared for example to lead oxide, in the preparation of optically transparent glasses. Previous attempts have lead to the formation of bismuth nanoparticles/crystals [18,19], opaque and dark-coloured glasses [4,18,20] and persistent crystallization upon the addition of very high or low bismuth content. [4] The most success has resulted from preparation of bismuth borates [1,4,5,7-9,21,22], silicates [6,19,20] and borosilicates [23] at melt temperatures below 1000 °C. Successful preparation of optically transparent bismuth boroaluminosilicate glasses has also been demonstrated [2]. The incorporation of Al_2O_3 was shown to reduce the likelihood of crystallization when added in limited quantities [2,4].

Investigation into Bi_2O_3 glasses has seen substantial progress over the past two decades, but the behaviour of bismuth cations in glass structure still remains difficult to characterize. ^{209}Bi NMR is impractical, save for some high-symmetry environments [24]. Vibrational

* Corresponding author.
E-mail address: jzwanzig@dal.ca (J.W. Zwanziger).

http://dx.doi.org/10.1016/j.jnoncrysol.2017.10.023
Received 12 September 2017; Received in revised form 11 October 2017; Accepted 12 October 2017
0022-3093/ © 2017 Elsevier B.V. All rights reserved.

spectroscopies including Raman [8,23,25,26] and IR spectroscopy [8,9,26] have been used to characterize Bi_2O_3-based glasses with some success, but cannot definitively determine the Bi-O coordination number. In addition, Bi_2O_3 can exhibit complex behaviour, acting as an intermediate between a network former and modifier [27] and having the ability to exhibit both multiple coordination and oxidation states [23].

Hence, this work examines the structural components present in a series of a bismuth boroaluminosilicate glasses with special consideration of the behaviour of Bi_2O_3. NMR spectroscopy is used for examining species with varying coordination numbers such as boron and aluminium. Raman spectroscopy is used to provide information regarding the nature of bismuth-oxygen interactions The behaviour of the experimentally determined stress-optic coefficient is compared with the predictions of the d/N_C model for various plausible Bi-O coordination numbers. We conclude that Bi_2O_3 can replace PbO to generate zero stress-optic glass, but that its efficiency in this regard is less than lead.

2. Methods

2.1. Sample preparation

Glass samples with a base composition of 86% B_2O_3, 10% SiO_2, and 4% Al_2O_3, and variable further addition of Bi_2O_3, were prepared, resulting in the nominal batch compositions listed in Table 1. The base glass composition was chosen following the work of Khanna et al., which showed that this composition leads to reasonably transparent bismuth-containing glasses [2]. Nominal compositions ranged from 20% Bi_2O_3 to 75% Bi_2O_3. The ten samples were prepared in 20 g batches from high purity Bi_2O_3 (99.9%), SiO_2 (99%), B_2O_3 (≥98%), and Al_2O_3 (≥98%). All reagents were obtained from Sigma-Aldrich and used without further purification. Mixed powders were melted in 95/5% platinum-gold crucibles in air at 850–1050°C for 1 h. Samples were poured into aluminum molds preheated to 300–450°C and annealed at the same temperature (just below T_g for the given sample) for 1–5 h in order to relax residual thermal stresses. Glasses were cut to obtain blocks with two sets of parallel faces. Opposing parallel faces were polished to approximately 1 μm surface roughness for optical measurements.

Compositions were verified through wavelength-dispersive spectrometry (WDS), performed on a JEOL JXA-8200 Electron Probe Micro-Analyzer operated at 15 kV and 20 nA and using a finely focused beam (10 μm). The four components present in the glass, Bi_2O_3, B_2O_3, SiO_2 and Al_2O_3, were analyzed in reference to the following standards: Bi_2S_3, $Li_2B_4O_7$ and CAM112 glass standard (for both Si and Al) respectively. Both nominal and experimentally determined compositions are reported in Table 1.

2.2. Density measurements

Sample densities were measured by the Archimedean method using ≥99% absolute ethanol as the immersion fluid. The density of absolute ethanol was corrected according to temperature for changes of 0.1°C or greater.

2.3. Stress-optic measurements

2.3.1. Stress-optic coefficient

Measurements of the stress-optic coefficient were made for each glass using the Sénarmont method [10,14,27,28]. Glass samples were subjected to compressive stress and induced birefringence was observed [14,28]. Each sample was loaded into a homemade stress gauge and situated on a light table. A white light source was passed through a linear polarizer which was then incident on the stressed glass and finally, observed through a second rotatable polarizer (analyzer). The analyzer was rotated to achieve θ, the angle at which transmission through the glass was at a minimum as estimated by eye. For each sample, multiple trials were conducted in which the stress was decreased incrementally and the corresponding θ was recorded. The error estimates listed are thus precision errors. The minimum intensity angle, θ, is directly related to the induced phase difference between the stressed and perpendicular axes, Δ, by $\theta = \Delta/2$. The phase difference is further related to the stress-optic coefficient, C, by

$$\Delta = \frac{2\pi d}{\lambda} C\sigma \tag{2}$$

where d is the thickness of the glass, σ is the applied stress, and λ is the wavelength of incident light. In SI units, C is given in Brewsters (10^{-12} Pa). An effective wavelength of $\lambda = 565$ nm was used, to approximate the light of the halogen tubes used in the light table (GE F8T5WW, 3000 K colour temperature).

2.3.2. Shear modulus

Values for the shear modulus, G, were obtained indirectly through the measurement of shear (transverse) velocity, v_T. To obtain v_T, ultrasonic pulses were passed through the glass, reflected off the back surface and received at the transducer in the form of an echo [27] using a Panametrics-NDT 25DL ultrasonic thickness gauge. The shear modulus is related to transverse velocity by:

$$G = v_T^2 \rho, \tag{3}$$

where ρ is the density of the sample.

2.3.3. Refractive index

Refractive index, n, was determined using a Woolam M-2000 variable angle spectroscopic ellipsometer. Polished samples were analyzed by single scan acquisition, at an angle of incidence of 55.0° and a range of wavelengths in the visible region, with the exact cutoff depending on

Table 1
Relevant data for d/N_C calculations. Both nominal and experimentally determined composition data are reported for bismuth boroaluminosilicate glass samples. Average boron and aluminum coordination numbers derived from the NMR studies are also listed for each sample. Uncertainties are in parentheses unless otherwise specified.

Nominal (mol %)					Experimental (± 3 mol%)					
x	Bi_2O_3	B_2O_3	SiO_2	Al_2O_3	Bi_2O_3	B_2O_3	SiO_2	Al_2O_3	n_{BO}	n_{AlO}
75	75.0	21.5	2.5	1.0	78.0	17.0	3.8	1.3	3.19(2)	4.4(1)
65	65.0	30.1	3.5	1.4	68.0	27.0	3.9	1.1	3.29(2)	4.2(1)
60	60.0	34.4	4.0	1.6	62.9	30.4	4.9	1.8	3.37(2)	4.4(1)
55	55.0	38.7	4.5	1.8	57.9	34.9	5.2	2.0	3.40(2)	4.4(1)
50	50.0	43.0	5.0	2.0	53.3	40.1	4.8	1.7	3.43(2)	4.4(1)
45	45.0	47.3	5.5	2.2	49.7	38.9	8.2	3.3	3.43(2)	4.5(1)
35	35.0	55.9	6.5	2.6	39.7	53.8	3.5	2.9	3.43(2)	4.7(1)
30	30.0	60.2	7.0	2.8	35.8	57.9	3.0	3.3	3.42(2)	4.7(1)
25	25.0	64.5	7.5	3.0	28.8	61.0	6.7	3.5	3.37(2)	4.8(1)
20	20.0	68.8	8.0	3.2	27.6	63.5	4.7	4.3	3.32(2)	4.8(1)

Download English Version:

https://daneshyari.com/en/article/7900375

Download Persian Version:

https://daneshyari.com/article/7900375

Daneshyari.com