Effect of antimony (III) oxide on reduction of bubbles from glass melting process

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Abstract. In this paper, studied the refining effects of Sb\textsubscript{2}O\textsubscript{3} and ultraviolet transmitting property of NaCaBSi glasses. These raw materials were weighted according to the (35-x)SiO\textsubscript{2}:30B\textsubscript{2}O\textsubscript{3}:15Na\textsubscript{2}O:10CaO:10ZnO:xSb\textsubscript{2}O\textsubscript{3} where x=0.0, 0.05, 0.1, 0.5, 1.0 and 2.0 mol%. NaCaBSi glasses were obtained by using melt quenching technique and characterized by using density, refractive index, optical absorption spectra and the number of bubbles in glass measure as a function of different concentrations. The density (ρ), molar volume (V\textsubscript{M}) and refractive index obtained were found to increase with increases in the concentration of Sb\textsubscript{2}O\textsubscript{3} in the glass matrix. The optical absorption spectra of glasses were measured in the wavelength of 200 – 2,000 nm. Number of bubbles in glass decrease when increasing Sb\textsubscript{2}O\textsubscript{3} concentrations.

Keywords: Sb\textsubscript{2}O\textsubscript{3}, NaCaBSi glasses, bubbles in glass, refining, molar volume

1. Introduction
This present, glass and the most common problem is the bubbles in the glass and focuses reduction of bubbles in the glass. Bubble in the glass is a crucial problem for glass manufacturers. Bubble gives optical distortion, which is detected and rejected easily by the customer. Countermeasures have been devised to reduce bubbles, but manufacturers have struggled with this unfinished problem worldwide [1-3]. The removal of gas bubbles is of great interest for many industrial processes. This is especially true for the glass industry where bubbles are formed by the physical trapping, within a highly viscous fluid, of atmospheric gases and the decomposition of raw materials. A classical method used to remove bubbles during glass melting is to introduce “fining” agents such as arsenic, antimony oxides or sulfate in small quantities [4]. The efficient melting factors defined in are temperature ensuring activity of the refining agent, glass composition (fining agent concentration, redox state of glass), reduced pressure, additional forces (ultrasonic, microwave, centrifugal) [5]. Antimony oxide, on the other hand, is in combination with antimony sulfide well used as a glass colorant and is known as “antimony ruby”. The creation of complex Sb\textsubscript{2}O\textsubscript{3}.Sb\textsubscript{2}S\textsubscript{3} occurs when the glass is reheated up to 700°C after casting [6]. The presence of cerium, antimony or arsenic oxide in a glass that already contains a coloring polyvalent ion may change the resulting color of such a glass. These oxides have reducing power that follows the order: Sb>As>S\textsubscript{n}>Cu>Ce. Therefore, the usage of these compounds should be considered individually, according to the present coloring oxide and its reaction to the oxidizing effect of the selected fining agent [7].

This research study the refining effects of Sb\textsubscript{2}O\textsubscript{3} and ultraviolet transmitting property of NaCaBSi glasses. The glass composition (35-x)SiO\textsubscript{2}:30B\textsubscript{2}O\textsubscript{3}:15Na\textsubscript{2}O:10CaO:10ZnO:xSb\textsubscript{2}O\textsubscript{3} where x is concentration of Sb\textsubscript{2}O\textsubscript{3} were synthesized.
2. Materials and Method

The current composition was prepared based on the proposed ratio: (35-x)SiO$_2$ :30B$_2$O$_3$ :15Na$_2$O :10CaO : 10ZnO: xSb$_2$O$_3$ where x=0.0, 0.05, 0.1, 0.5, 1.0 and 2.0 mol%. The main chemical composition was SiO$_2$ which was obtained from local sand to prepare glass. All the chemicals were weighted according to the stoichiometric amount and thoroughly mixed. The powder were mixed well was added to porcelain crucible and melted at 1200ºC for 3 h using a furnace. The melt glass samples were obtained by pouring on graphite mold. The obtained transparent glass samples were finally annealed at 500ºC for 3 h. The furnace temperature is reduced to the room temperature with a cooling rate of 10ºC/min to reduce thermal stress and cool down to the room temperature. Finally, the good qualities of clear and transparent glasses were formed. Fig.1. Shows the photographs of glass samples.

![Figure 1. Digital photograph of Sb$_2$O$_3$ doped in glasses](image)

The glass sample densities were determined by using Archimedes method Eq. (1). Water was selected as an immersive fluid. For all samples, the measurement was done at room temperature and repeated three times to reduce the error.

$$\rho = \frac{w_A}{w_A - w_B} \times \rho_{ref}$$  \hspace{1cm} (1)

Where $w_A$ is the weight of the glass sample in air, $w_B$ is the weight of the glass sample in water and $\rho_{ref}$ is the density of water at room temperature (27ºC), which is 1 g/cm$^3$. Consequently, the molar volumes of the glass samples were calculated based on the density values according to the following equation:

$$V_M = \frac{M_T}{\rho}$$  \hspace{1cm} (2)

Where, $V_M$ is the molar volume, $M_T$ is the total molecular weight and $\rho$ is the calculated density of the glass. Absorption spectra were recorded by using UV-VIS NIR Spectrophotometer (Shimadzu UV-3600), working in 200 – 2,000 nm at room temperature. The refractive indices were measured by using an Abbe refractometer (ATAGO) with a sodium vapor lamp as a light source having wavelength of 589.3 nm (D line) with Monobromonaphthalene as a contact layer between the sample and prism of the refractometer [8, 9]. The bubbles in glasses Measured by Digital microscope.

3. Results and discussions

The density of the materials is an important quantity of the physical parameters to determine the change in softening/compactness of the structure, change in geometrical configuration, coordination number, cross-link density and, the dimension of interstitial spaces of the glass. The density of glasses increasing with the increasing of Sb$_2$O$_3$ concentration.
The molar volume is almost stable from 0.0-0.1 mol% concentration and increased when concentration is in the range of 0.1-2.0 mol%, due to Sb\(^{3+}\) entering into the glass network as a modifier and non–bridging oxygen as shown in Figure 3.[10]

**Figure 3.** Molar volume of glass sample

The refractive index of glass samples increases with increase in the concentration of antimony oxide, this is due to the increase in density of the prepared glass samples (fig.4.). When the density of the developed glass samples increases the structure of the glass will be compact and velocity of light retards from glass which causes the refractive index to increase as shown in Figure 4.

**Figure 4.** Refractive index of glass samples
The bubbles in glass measured by digital microscope were shows bubbles in glass decreases tend with increasing of Sb$_2$O$_3$ concentration as show in Figure.5.

![Figure 5. The bubbles in glasses (A-F)](image)

The absorption spectra of Sb$_2$O$_3$ doped glasses obtained by UV-visible spectrophotometer are illustrated in figure 6. The absorption peak was found at 1,050 nm which is Fe$^{3+}$ the highest peak is 0.0 mol% and decreases with increasing of Sb$_2$O$_3$ concentration because of oxidation change between Sb$_2$O$_3$ and Fe$_2$O$_3$ in the glass as show in Figure.6. The charge transfer of electrons from the Fe to the Sb is showed as redox eq. (3) [11].

$$Fe^{2+} + Sb^{3+} \rightarrow Fe^{3+} + Fe^{2+}$$  (3)

The changing between the Fe$^{2+}$ to Fe$^{3+}$ ions was occured. The emergence of light yellow color is a combination of Fe$^{2+}$ and Fe$^{3+}$ ions in glass [12].

![Figure 6. Absorption spectra of glass samples](image)
4. Conclusion
The study of the bubble in glass found to increase of Sb$_2$O$_3$ concentrations causing the bubbles in the glass to decrease. Density and refractive index of glass increase with increasing of Sb$_2$O$_3$ concentration. The bubbles in glass measured by digital microscope was shows bubbles in glass decreases tend with increasing of Sb$_2$O$_3$ concentration. The absorption peak was found at 1050 nm which is Fe$^{3+}$ the highest peak is 0.0 mol% and decreases with increasing of Sb$_2$O$_3$ concentration because there is an oxidation change between Sb$_2$O$_3$ and Fe$_2$O$_3$ in the glass.

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