Effect of Sintering Temperature on Silica Based Glass-Ceramics Derived From Soda-Lime-Silica Glass

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Abstract: Silica-based glass-ceramics have been fabricated in this work using soda-lime-silica (SLS) glass waste. The quarternary glass-ceramic was fabricated with composition xMgO: (0.5-x)ZnO: (0.2)B_2O_3: (0.3)SiO_2 ; x = 0.01, 0.03 and 0.07 mole using melt quenching technique. The glass samples were sintered at 27°C, 700°C and 800°C. The variation composition of MgO on physical properties which includes density measurement and linear shrinkage measurement had been studied. The density of glass samples that are treated at room temperature and 700°C is almost having the same values which about 1.8g/cm^{-3} for different percentage of MgO. Then, density for samples that are treated at 800°C had decreased from 1.855g/cm-3 to 1.658g/cm^{-3} and increased again to 1.815g/cm-3 for different percentage of MgO. Linear shrinkage of glass samples with variation composition of MgO at room temperature is zero because the samples are not heated which have no decreasing in diameter. The samples that had been treated at temperature 700°C and 800°C had increased and decreased in increasing the composition of MgO.

Keywords: Sintering, Silica glass, Glass ceramics, physical properties, linear shrinkage.

Soda-lime glass, also called soda-lime-silica (SLS) glass, is the most prevalent type of glass, used for windowpanes and glass containers (bottles and jars) for beverages, food, and some commodity items. Glass bakeware is often made of borosilicate glass. Soda-lime glass accounts for about 90% of manufactured glass. Soda-lime glass is relatively inexpensive, chemically stable, reasonably hard, and extremely workable. Because it can be softened and remelted numerous times, it is ideal for glass recycling. It is used in preference to chemically-pure silica, which is silicon dioxide (SiO_2), otherwise known as fused quartz.

The study on silica-based glass-ceramic derived from SLS glass needs to be performed to make the glass-ceramic which can be used in Low-Temperature Co-fired Ceramics (LTCC) technology. In the normal LTCC technology, ceramic green tapes are processed by punching and screen printed to make vertical interconnect and coplanar conductor patterns, laminated and eventually fired at 850 ºC to make an extremely integrated substrate. The low sintering temperature provided by the LTCC technology is that the key factor enabling its advantageous utilization for today packaging ideas in microwave modules [1]. However, the literature has not reported the utilization of the SLS glass waste in the fabrication of silica-based glass-ceramics. The SLS glass waste provides the required SiO_2 and these wastes are expected to provide another alternative means of managing the disposed of SLS glass wastes in the metropolitan cities around the world. This work studies the physical properties of the fabricated silica-based glass-ceramic through XRF, and FTIR characterizations. The work also studies the effects of the temperature of sintering on the physical and phase analysis of the silica-based ceramics.

Nine glass-ceramic pellets sample of composition xMgO: (0.5-x)ZnO: (0.2)B_2O_3: (0.3)SiO_2 ; (x =1, 5, and 7 % of mole) were prepared by using melt quenching technique.

Appropriate 13g composition of MgO, ZnO, B_2O_3 and SiO_2 were weighed using electronic balance having accuracy 0.0001g. The calculation of mass and that use in research and the actual mass has been used is shown in the Appendix. Then mix them together by using agate mortar for 5 to 10 minutes. The mixture of composition then is placed in the alumina crucible and melt it in the furnace at temperature7 1200°C for 3 hours 50 minutes. The melted glass was decanted into a pail of purified water to gain frits of glass. Then they were ground into a soft powder and 4 scoops of spatula of polyvinyl alcohol (PVA) are added into the soft powder which played a role the binder to gain the pellets for using them to study the characterization. The soft powder was compressed using hydraulic jack at an applied load of 3200 psi at interval 2 and a half minutes between 1600 psi. The thickness and the diameter of the pellets were measured using vernier calliper before the heat treatment. The pellets have been heated at temperature 700°C, 800°C and 1000°C for 1 hour 20 minutes, 1 hour 37 minutes and 1 hour and 50 minutes respectively. Finally, the thickness and diameter of the pellets were measured again.

Table 1: The physical properties of xMgO: (0.5-x)ZnO: $(0.2)B_2O_3$: $(0.3)SiO_2$ glass sample

MgO %	Density (g/cm^3) at different temperature (±0.01 °C)			OPD (g-mol/l) at different temperature (±0.01 °C)			Molar Volume (cm^3)		
	Control	700 °C	800 °C	Control	700 °C	800 °C	Control	700 °C	800 °C
1	1.809	1.813	1.836	175.310	175.742	177.935	39.929	39.831	39.340
3	1.841	1.854	1.902	180.505	181.702	186.415	199.857	206.644	209.519
7	1.992	2.059	2.088	38.780	38.525	37.551	35.025	33.875	33.410

The density of samples was determined by using Archimedes' principle and this density test was carried out at room temperature. All samples were in the form of pellet before undergoing the density measurement and distilled water was used as the buoyancy liquid. The density of the pellet was determined according to the Archimedes principle and calculated by using the formula:

$$\rho = \frac{W_a \rho_b}{(W_a - W_b)} \quad (1)$$

where W_a is the weight in the air, W_b is the weight in distilled water and ρb is the density of distilled water, which is 1.0 g/cm^3. The unit for density is gram per centimetre cubic (g/cm^3). The glass network compactness was related to the density of glass system with affected by the structural softening or compactness, the coordination number is changing and the dimension of interstitial spaces of glass [2]. Besides, the relationship between density and molar volume is inverse to each other in behaviour, but, in the present glass system, the relationship is different [3]. The oxygen packing density and molar volume is calculated by using glass density relation following equations:

$$\text{Oxygen packing density} = ((1000 \times \rho \times 0))/M \quad (2)$$

$$\text{Molar Volume} = M/\rho \quad (3)$$

where ρ = mass density, M= Molecular weight of glass composition and O = number of oxygen atoms in the composition [4]. The density of glass-ceramic depends upon the various factors such as crystallisation temperature, cooling rate and phase formation [5]. Besides, increasing the temperature or duration of the heat treatment process significantly increases the linear shrinkage of the materials. The compact disk sample's linear shrinkage depends on temperature changes. The increase in the percentage of linear shrinkage is due to the reduction in the sample's total fractional porosity with the increase in temperature for heat treatment. Wollastonite glass-ceramics can have an increase or both increase and decrease in percentage value with an increase in sintering temperatures [6]. Furthermore, with the increase in temperature or duration of the heat treatment process, the linear shrinkage of the materials increases significantly.

The density measurement that has been carried out showing that the sample that is not being sintered (room temperature) has the lowest density for all percentage of MgO. The samples that have been sintered at 700 ℃ have slightly increased in terms of the density. The increment pattern continuously towards the MgO percentage of 7%. The density increased at range from 1.809 g/cm^3 to 2.088 g/cm^3 from 1% mole to 7% mole of MgO. The density of the glass structure evaluates the compactness of the glass structure and the changes that might be due to changes in phase. The decrease in molar volume and an increase of the density was due to the molecular weight of MgO and ZnO. The atomic number and radii of Mg^+ are lower than Zn^+, and Si^{2+}. Therefore, it indicates that the Mg^+ located in between the interstitial of the atomic. The decrement of molar volume also indicated the decrement in the bond length or an interatomic spacing between atoms. The oxygen packing density was increased at range 175.310 g-atom/l to 209.519 g-atom/l as MgO content increases in the glass structure as illustrated in Figure 4.3. These can be explained from a structural point of view. The structure becomes less packed with increased in mole percentages of MgO in the molecular interstitial. Less non-bridging oxygen atom might also occur as a percentage of MgO increased due to the increment of Mg^+ in the glass structure.

Linear shrinkage is the percentage of decrease in ceramic sample length and it is of great importance in the ceramic industry. A few applications, the length is required to determine the volume of the produced ceramic sample and its dimensions before the formatting process. Linear shrinkage is inverse to the particle radius but is not greatly affected by sintering time. It is also due to the composition and tends to increase as the fluxing components increase with the production of relative phases during the firing. Length of produced samples was measured before and after sintering. Linear shrinkage defines the change in pellets dimension with sintering. Determination of the linear shrinkage can be made by measuring the initial dimension of the pellet before sintering and after sintering and applying the values in the following equation;

$$\text{Linear shrinkage (\%)} = [(L_o - L) \div L_o] \times 100\% \quad (4)$$

where L_0 and L are the initial dimension and dimension after sintering respectively [7].

Referring to Figure 1, linear shrinkage for all mole percentage of MgO was decreased when the sintering temperature was increased. Non-sintered glass samples have zero value because there are no changes in the diameter of the pellets. The linear shrinkage for glass samples that had been sintered at temperature 700 °C was decreased and increased again through increasing the composition of MgO. But for the glass samples that had been sintered

at temperature 800 °C, the linear shrinkage starts to increase and decreased again when the composition of MgO increased.

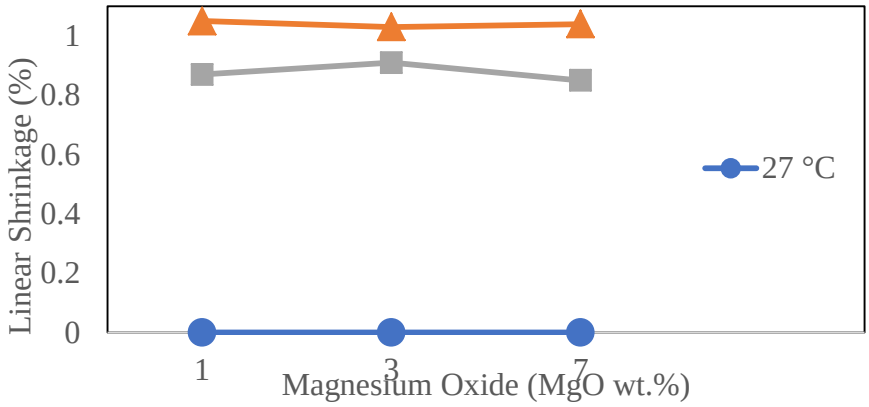

Figure 1: Linear shrinkage variation with fractional compositions of magnesium oxide (MgO) for various sintering temperatures of silica-based glass-ceramics.

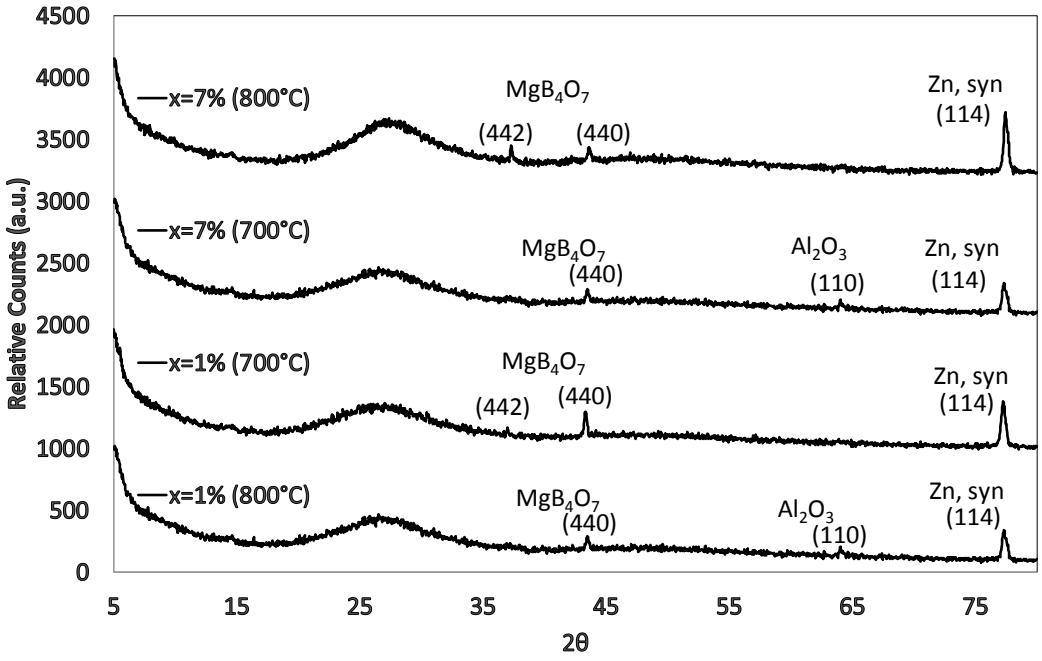

Figure 2, X-ray diffractogram phase of xMgO: (0.5-x)ZnO: (0.2)B_2O_3: (0.3)SiO_2 glass sample for x = 0.01, 0.03 and 0.07 (JCPDS reference code: 01-076-0938).

XRD analysis was performed using a Bruker-D8 X-ray diffractometer to confirm the amorphous nature of the prepared glass samples. An absence of sharply defined peaks which confirms the nonexistence of long-range order and therefore the amorphous nature of the prepared glasses. Based on Figure 2, it illustrated the X-ray diffractogram phase, which indicates an amorphous state of xMgO: (0.5-x)ZnO: (0.2)B_2O_3: (0.3)SiO_2 glass sample for x = 0.01, 0.03 and 0.07. The appearance of the broad hump at a 2θ angle of 27° until 30° indicates that the overall structure was an amorphous state with the presence of crystalline

phase due to sintering treatment towards the samples. It is proven that the reaction between the network modifier and the network intermediate results in the forming of MgB_40_7. Nevertheless, it is also observed that the XRD diffractogram also indicate the peaks for $Al2O_3$ which is from the glass bottle and crucible.

In conclusion, the nine samples of ternary glass-ceramic were successfully prepared using SLS glass waste as a source of silica in the $MgO\text{-}ZnO\text{-}B_2O_3\text{-}SiO_2$ glass system. The density that obtained from glass-ceramic samples was almost the same which is about 1.8 g/cm^3 for non-sintered samples and samples that were heated at temperature 700 °C. For samples that were treated at temperature 800 °C, the density was decreased from 1.855 g/cm^3 to 1.658 g/cm^3 and its increased to 1.815 g/cm^3 when the mole percentage of MgO was increased. Besides, the linear shrinkage of the non-sintered glass sample is zero for all composition because there are no changes in the diameter of the pellets. The glass samples that had been heated at temperature 700 °C was decreased from 1.05cm to 1.03 cm and continue increased from 1.03 cm to 1.04 cm when the mole percentage of MgO increased. For glass samples that had been sintered at temperature 800 °C, the results were inverse from results obtained for glass samples sintered at temperature 700 °C. The results increased from 0.87 cm to 0.91 cm and decreased back to 0.85 cm when the mole percentage of MgO increased. Furthermore, the X-ray diffractogram phase, which indicates an amorphous state of $xMgO: (0.5\text{-}x)ZnO: (0.2)B_2O_3: (0.3)SiO_2$ with the appearance of crystalline phase due to sintering treatment towards the samples which indicate the formation of MgB_40_7.

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