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PREPARATION AND CHARACTERIZATION OF FOAM GLASS FROM SODA LIME SILICATE GLASS WASTE BY USING DIFFERENT DOSAGES OF LIMESTONE

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Abstract

Soda-lime silicate (SLS) waste glass is a kind of solid waste that is produced in large quantities as container glass or flat glass. In this work, waste window glass is used to produce foam glass using limestone as a foaming agent. The pore structure of the foam glass and the optimization of its properties by adjusting the amount of glass and foaming agents were investigated in this work. The formation of foam glass was studied on samples prepared from blends of SLS waste glass with 0.5 wt% SiC and 2, 4, 6 wt% limestone by sintering at 800 °C. The foam glass with 2wt % limestone presented a homogeneous foam structure with low body density (0.26 g/cm^3), apparent porosity (9 vol%) and thermal conductivity (0.04 W/m·K). When the limestone content increases to 6 wt %, the density and thermal conductivity of the sample also increases to 0.92 g/cm^3 and 0.13 W/m·K, respectively.

Keywords: *foam glass, foaming agent, glass waste, limestone*

1. Introduction

Soda –lime silicate glass is a transparent frozen liquid of calcium carbonate, soda ash, and silica that has been melted at high temperatures. The melted liquid cools rapidly and the high viscosity prevents from crystallization (Vieitez et al., 2011; Bauchy and Micoulaut, 2015). The main component of silicate glass is silicon dioxide (SiO_2), which is extracted from sand (Butler and Hooper, 2019). Waste glass can be recycled into industrial foam glass, which can reduce the amount of waste glass and greenhouse gas emissions to the atmosphere by applying it as an insulation material (König et al., 2015).

Foam glass, an advanced type of environmentally friendly material usually produced from waste, has a porous structure. The type of pores determines the properties and applications of foam glass. Foam glass is a kind of fire and moisture resistant, lightweight, high strength construction and decoration material with high thermal insulation performance and sound absorption. It has a density range of 0.4-1.0 g/cm^3. Closed-cell foam glass, in which the open porosity is less than 10 vol%, is a construction material that outperforms typical thermal insulation materials in terms of low thermal conductivity,

compressive strength of more than 0.5 MPa, water resistance, and long service life. When used in subway stations and gardens, foam glass with an open porosity of more than 50 vol% provides high sound insulation and low water absorption (Ogundairo et al., 2019; Gopi, 2009). In general, any glass can be formed into foam with a suitable foaming agent such as silicon carbide, limestone, carbon black, manganese oxide, gypsum and etc.

Limestone decomposes at temperatures around to 700-900°C, producing large amounts of gas, and therefore forms mainly an open-pore structure (Karazi et al., 2017; Petersen et al., 2014). The composition of the released gases depends on the foaming agent and the composition of the pristine glass. The gases produced during the decomposition of carbonates or sulphates consist mainly of CO_2 and SO_2, respectively. The aim of this paper is to study the effect of limestone dosage on the foaming process and the foam glass properties.

2. Experimental procedures

2.1. Raw materials

The raw materials used in this work to produce foam glass were soda-lime silicate glass (SLS), obtained from window glass waste; limestone, a carbonate sedimentary rock ,the limestone powder was obtained from the Hungarian SZIKKTI laboratory (coded as limestone 0.0); and silicon carbide. Waste glass and limestone were crushed separately and dry milled for 20 minutes at 170 rpm in a planetary ball mill (RETSCH PM 400) with silica balls.

2.2. Techniques for the characterization of raw materials

The composition of the limestone was studied by phase identification using the Rigaku Miniflex II X-ray diffractometer (XRD). A CILAS 715 Granulometre instrument was used to determine the particle size distribution of all raw materials. The tests were done in water, as a liquid medium, sodium-tripolyphosphate as a dispersing agent and 1 minute of ultrasonic treatment was also used. The SLS and limestone were ground and sieved to a particle size of 100 μm, and the particle size distribution of the powders was measured. Camar Elettronica heating microscope was used for thermo-optical analysis of SLS glass. The following specific values can be determined:

A. Sintering: in which local melting occurs, preferably at the particle contacts. This process results in shrinkage of the sample while maintaining its original shape. The starting point is sometimes defined as the point at which the height of the sample falls below 95% of its original value (Heo et al., 2012).
B. Softening: at which the first signs of softening of the sample, such as rounding of the edges or disappearance of the unevenness of the sample, become visible.
C. Sphere: at which the shape of the sample forms a sphere and the height of the sample is equal to its width.
D. Hemisphere or half sphere: at which the shape of the sample forms a spherical cap and the height of the sample is typically half the length of its base or 2/3 of its original height.
E. Melting: at which the sample spreads out on the substrate and forms a layer with a thickness that is 1/3 of its original height.

The tests were done on pressed cylindrical specimens. Thermal analysis, including thermogravimetric analysis (TG), differential thermal analysis (DTA) and differential thermogravimetric analysis (DTG) of limestone powder was performed by used the MOM

Derivatograph C. The bulk density of the glass powder was measured by the ratio between the mass (M) and the volume (VF) occupied by the powder.

2.3. Sample preparation and test methods

Powder mixtures were prepared from SLS glass by adding 2, 4, 6 wt% limestone and 0.5 wt% of silicon carbide sieved at < 100 μm, as shown in ***Table 1***.

Table 1. Sample composition (in wt%)

Sample code	SLS glass	SiC	Limestone
GL2	97.5	0.5	2
GL4	95.5	0.5	4
GL6	93.5	0.5	6

The powder mixtures were prepared by mixing in a planetary ball mill. The milling conditions were always the same. Then, the powder mixtures were pressed into cylindrical sample with a diameter of 20 mm at a pressure of 140 MPa using a single-axis pressing machine. The prepared specimens were sintered for 10 minutes at 800 °C in a programmable high-temperature furnace at a heating rate of (5 °C/min). After sintering, the specimens were left in the furnaces and cooled to room temperature.

2.4. Techniques for the characterization of foam glass samples

The powder mixtures were analyzed with a heating microscope. The change in sample height as a function of temperature was recorded. Microstructural of the glass foam were performed using optical microscope. The volume expansion of the glass foam was determined by measuring the volume of the sample before foaming (V_0) and the volume of the sample after foaming(V_1) according to equation (1):

$$V = \left(\frac{V_0 - V_1}{V_1}\right) \cdot 100;\ [\%] \tag{1}$$

The density of the foam glass samples was determined by measuring their weight and dimensions. The apparent porosity was evaluated according to equation (2), where P_0 is the apparent porosity (vol%), d_a is the apparent density, and d_b is the density of the glass waste.

$$P_0 = 1 - \left(\frac{d_a}{d_b}\right) \cdot 100;\ [\%] \tag{2}$$

Water absorption was measured according to ASTM C373-88 (Standard ASTM C373-88: 1999). This involves the drying of the samples to a constant weight (D) and soaking them in distilled water for 24 hours at room temperature. The test was performed on five representative specimens. Water absorption, WA (wt%), expresses the relationship of the weight of water absorbed (M) to the mass of the dry specimen (D) as follows:

$$WA = \left(\frac{M - D}{D}\right) \cdot 100;\ [\%] \tag{3}$$

3. Results and discussion

3.1. Characterization of raw materials

Figure 1 shows the X-ray diffraction pattern of limestone. The peaks show the chemical composition of the limestone Mg 0.03 Ca0.97 (CO3).

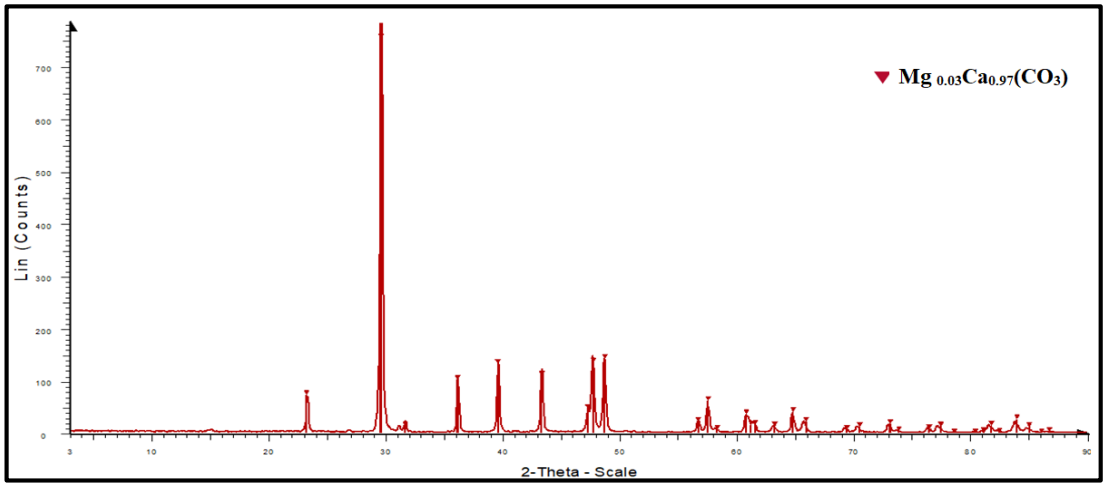

Figure 1. *X-ray diffractogram of limestone*

The characterization of the initial particle size distribution is an essential step in the foam production, as the pore diameter of the resulting material depends on the fineness of the initial powders. Figure shows the particle size distribution curves. It can be concluded that limestone powder was fine-grained, 12 % of the particles are less, than 1 µm, the average particle size 6 µm. SLS glass is coarser than limestone. It contains submicronal particles only in traces. The average particle size of SLS glass is 45 µm.

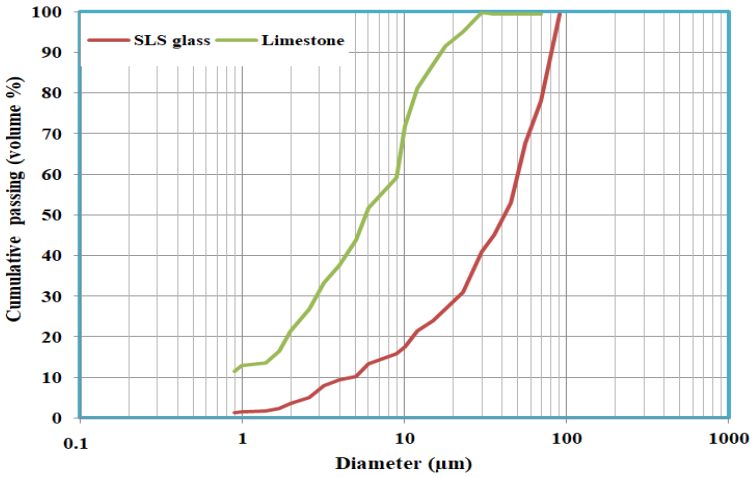

Figure 2. *Particle size distributions of the milled and sieved raw materials*

Heating microscopy can be used to characterize the melting behaviour and observe phenomena occurring in situ, as well as to determine the characteristic temperatures of a material. The heating microscope shows temperature levels of SLS glass: the sintering temperature at 668 °C, the softening point at 780 °C, the sphere point at 953 °C, hemisphere point (half sphere) at 993 °C and the flow point (melting) at 1042 °C, as shown in Figure .

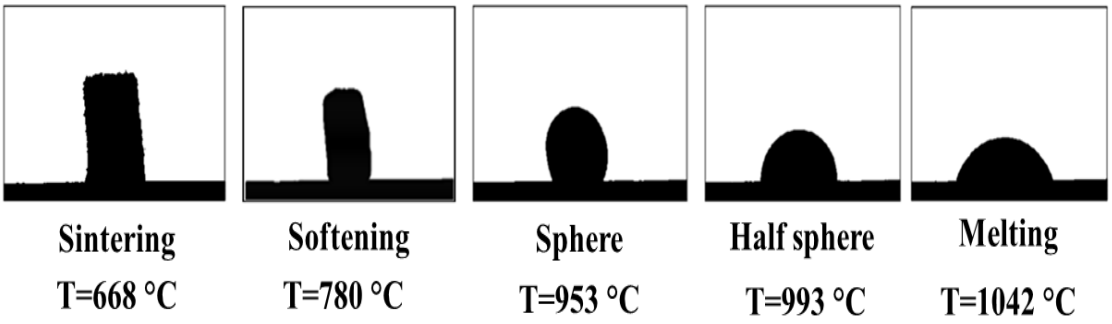

Figure 3. *Heating microscope images of SLS glass*

3.2. Characterization of foam glass

Thermogravimetric analysis (TGA) measures the weight changes of a material as a function of temperature (or time) under a controlled atmosphere. The TGA of limestone was measured up to the specified decomposition temperature and the mixtures were tested with a heating microscope to determine the effects of the limestone dosages as on the foaming temperature, and maximum height of the samples during heating as shown in Figure 4.

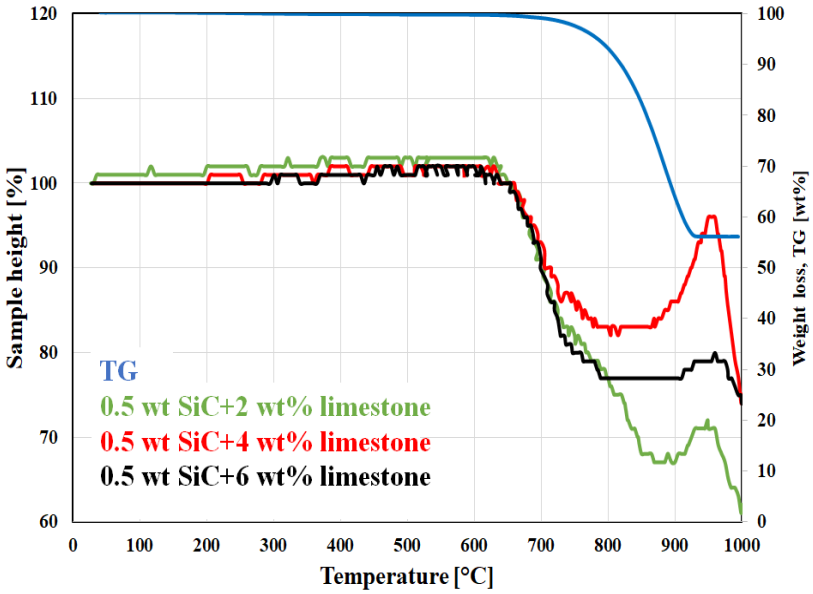

Figure 4. *Thermal decomposition of limestone and foaming curves of the glass mixtures having 0.5 wt% SiC and 2, 4, or 6 wt% limestone*

For a successful foaming process, it is important that the gasses, released by the foaming agent(s), are entrapped in the sintered body. At an elevated temperature, calcium carbonate (calcite) decomposes to solid calcium oxide (CaO, lime) and gaseous carbon dioxide (CO_2) according to the following one-step chemical formula (García-Ten et al., 2012; Venkateswarlu et al., 2014):

$$CaCO_3(s) \rightarrow CaO(s) + CO_2(g) \quad (4)$$

Calcium oxide formed during the decomposition (endothermic reaction) of the limestone at the temperature range of 700-930 °C (Figure). The initially slow process accelerates at 750 °C and the rapid change indicates an increased transformation rate beyond the temperature of 750 °C. The appearance of the calcium oxide and CO_2 phases starts at about 700 °C and lasts up at about 930 °C, denoting the completion of the thermal decomposition (Kohobhange et al., 2019). One of the most important factors for foaming is the viscosity of the mixture. When the viscosity becomes inadequate during foaming, the gas can escape through the surface of the pressed compacts into the atmosphere. As can be seen in Figure the volume expansion of the foam sample is inversely proportional to the limestone content.

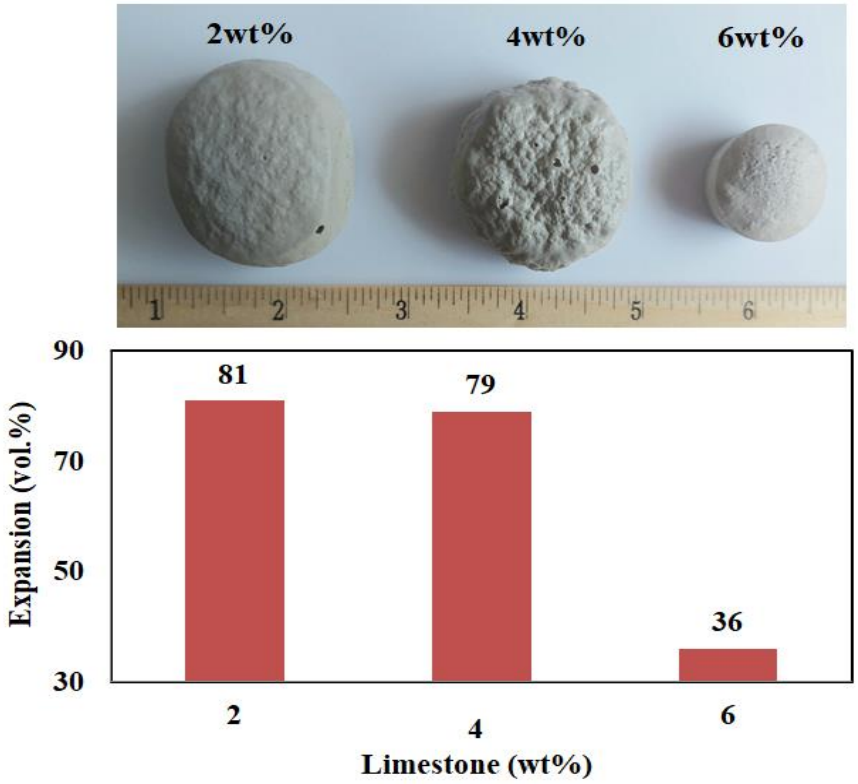

Figure 5. *Volume expansion of foam glass with 2, 4, and 6 wt% limestone content*

Since the surface layer containing the completely decomposed carbonate becomes thinner and the gas can escape faster and more freely, increasing the amount of limestone can cause the shell to decompose. (König et al., 2014). Therefore, the size hardly changes when the limestone content is up to (6 wt%) shows the optical microscopy images of the samples in Figure 6.

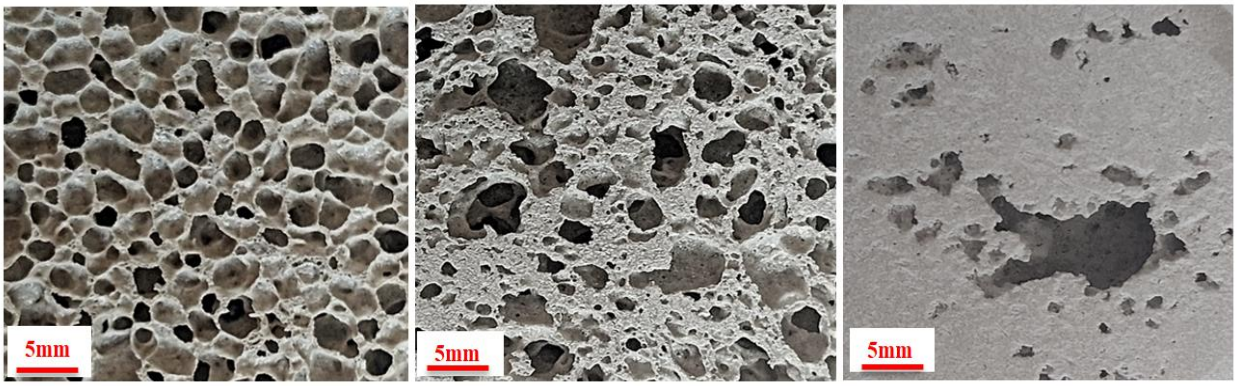

Figure 6. *Microstructure of the SLS foam glasses with (a) 2 wt%, (b) 4 wt%, (c) 6 wt% limestone (scale bar =5mm)*

The sample with 2 wt% limestone had a noticeably cellular microstructure, with a quite homogeneous cell size distribution and fairly thin cell walls. Significant parts of the cells were closed. The pore size distribution of the sample with 4 wt% limestone was not homogeneous; the structure is denser. The structure is more porous than cellular, as interconnected cells and cell junctions are hardly found in the microstructure. As for the sample with 6 wt% limestone content, the formation of small pores is only visible in the inner cross section; otherwise the structure is quite dense.

The results of the bulk density and apparent porosity test are shown in the Table 2.

Table 2. *Bulk density and apparent porosity results*

Sample code	**GL2**	**GL4**	**GL6**
Bulk density (g/cm^3)	0.26	0.28	0.92
Apparent porosity (vol%)	91	90	70

The density and apparent porosity of the sintered SLS foam glasses is in the range of 0.26-0.92 g/cm^3 and 70-81 vol%, respectively. The bulk density increases while the apparent porosity decreases with the limestone content.

Normally, the thermal conductivity depends on the chemical composition, cell size and wall thickness of the foam. The thermal conductivity decreases as the size of the cells increases (Sassi and Simon, 2022). The cells are able to block the transmission of the heat flow. The heat flow changes direction when it hits the cell and spreads through the wall. The thickness of the wall affects the thermal resistance; the thicker the wall, the lower the thermal resistance. Thermal insulation can be improved by better distribution and arrangement of the cells (Sassi et al., 2020). The results of thermal conductivity are shown in Figure 7 it was found that the thermal conductivity of the glass foam increases when the dosage of the limestone increases due to the increase in density.

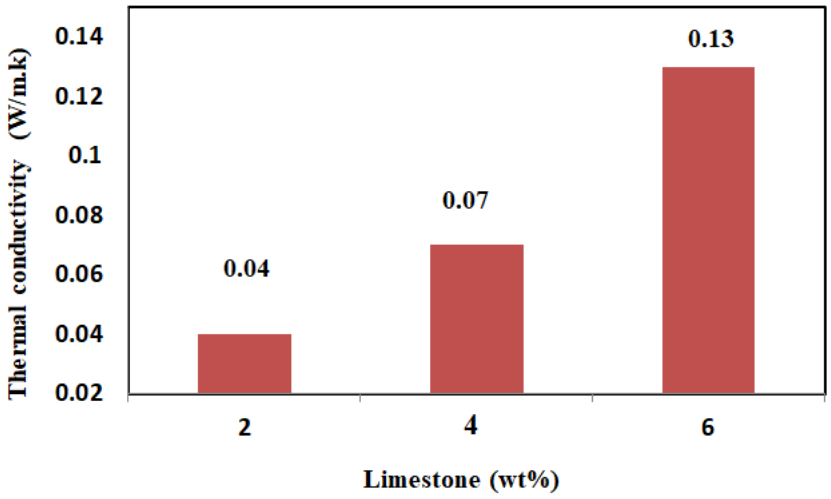

Figure 7. *Thermal conductivity of the SLS foam glasses with 2, 4, or 6 wt% limestone content*

4. Conclusion

The production of foam glass is a method of recycling glass waste and has many applications. Foam glass was made from waste window glass, with SiC and limestone as foaming agents. The aim of this work is to investigate the influence of limestone in different dosages on the production of glass foam . The foams with 2 wt% limestone content exhibited acceptable bulk density (0.26 g/cm^3) and thermal conductivity (0.04 W/m·K) with a homogeneous, cellular structure. As the limestone content increases to 6 wt%, no cellular structures are developed, possibly because a large amount of CO_2 gas is released and coalescenced, leaving the softened glass through its shell.

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