THERMAL INSULATING GLASS FOAM WITH HIGH MECHANICAL STRENGTH MADE BY MICROWAVE RADIATION

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ABSTRACT: The paper presents results of research in the manufacturing domain of some glass foams with thermal insulating characteristics (apparent density of 0.50 g/cm^3, porosity of 76.2% and thermal conductivity of 0.149 W/m·K) and with a very high compressive strength (16.6 MPa). The product was achieved by sintering at 853 ºC of a pressed mixture composed from 78.2% container glass waste, 1.0% sodium carbonate, 2.0% glycerol, 7.8% sodium silicate and 11.0% water. The adopted heating technique was the predominantly direct microwave heating in a 0.8 kW-microwave oven, which allowed to obtain a low specific energy consumption of 1.10 kWh/kg.
KEYWORDS: glass foam, microwave heating, high mechanical strength, thermal insulating material, efficiency energy.

1. INTRODUCTION

In the last decades, the waste recycling (metal, plastic, glass, paper, etc.) has become an important concern of the civilized world both economically and especially, ecologically. The waste commonly comes from post-consumer container glass and window glass from demolition and rehabilitation of building. The annual generation rate of these types of waste is constantly increasing. Part of it is reused as a raw material in the industrial production of the new glass. But the glass waste processing for the manufacture of the new glass involves high costs for the selection of the colored cullet. Storing glass waste in landfills would require large areas of land used irrationally. In addition, the danger of soil contamination there is. An efficient and useful solution proved to be the foaming of glass waste by a heat treatment at high temperature and obtaining porous and light products with low thermal conductivity and sufficiently high mechanical strength, suitable for their use as an alternative to some existing construction materials.

The objective of the works presented in the paper is to experimentally manufacture a light glass foam with a high mechanical strength in the conditions of achieving a low specific energy consumption.

A dense glass foam with an apparent density between 0.60-0.90 g/cm^3, a thermal conductivity between 0.081-0.105 W/m·K and a compressive strength between 2.5-6.2 MPa was produced by the technique of mixed microwave heating of a pressed powder mixture of 90-94% colorless glass waste, 5% sodium borate (borax), 1.5-5.0% calcium carbonate and 8.5% water addition. The temperature of the sintering/foaming process was between 820-851 ºC. The specific energy consumption had values between 1.40-1.74 kWh/kg [1].

Geocell Schaumglas (Austria) is one of the leading European manufacturers of foam glass gravel. The ra material is 90% colored post-consumer container glass and 10% colorless window glass waste. The foaming agent is not mentioned. The process temperature is below 900 ºC. The foam glass gravel has a porous structure, the bulk density being of 0.15 g/cm^3. The thermal conductivity is of 0.08 W/m·K and the compressive strength is of 5.7 MPa [2].

The Swiss company Misapor Switzerland produces foam glass gravel of 98% recycled glass, 2% gypsum, limestone or silicon carbide as a foaming agent individually used. The average process temperature is of 900 ºC. The main characteristics of the product are: apparent density between 0.21-0.25 g/cm^3, thermal conductivity of 0.12 W/m·K and compressive strength between 4.9-6.0 MPa [2].

Glapor Werk Mitterteich (Germany) is also specialized to manufacture foam glass gravel. The powder mixture includes 87% recycled glass (flat glass or container glass waste), 1% glycerol, 12% sodium silicate and below 0.5% kaolin. The bulk density of the glass foam is between 0.13-0.21 g/cm^3 and the compressive strength is in the range 4.9-6.0 MPa. Using a liquid foaming agent (glycerol) the

porosity of the pieces is very fine with the pore size below 300 μm [2].

The foam glass gravel industrially made uses conventional heating techniques and the specific energy consumption of these industrial processes is not mentioned in literature.

Several experiments on the manufacture of foam glass gravel in a 0.8 kW-microwave oven in the Romanian company Daily Sourcing & Research highlighted the optimal sample achieved of 83% glass waste, 1% glycerol, 8% sodium silicate and 8% water addition. The temperature process was 823 ºC and the specific energy consumption was very low (0.88 kWh/kg). The main characteristics of the optimal sample were: apparent density of 0.24 g/cm^3, thermal conductivity of 0.063 W/m·K and compressive strength of 5.9 MPa almost similar to those industrially obtained [2].

The paper [3] presents results of experiments on manufacturing glass foams with excellent compressive strength. The composition of the starting materials for the manufacture of the best sample included 92% container glass waste, 3.5% sodium silicate as well as 3.5% yellow glycerol and 1% sodium carbonate as foaming agents. Yellow glycerol was obtained from the production of biodiesel containing 80% glycerol, 10% water, 10% methanol, fatty acids and other materials). The pressed powder mixture was heated to 850 ºC for 30 min, with an average heating rate of 10 ºC/min. The used heating technique was the conventional method. The bulk density had the value of 0.67 g/cm^3, the thermal conductivity was 0.260 W/m·K and the compressive strength reached 16 MPa.

Another porous material with high mechanical strength was manufactured in the microwave field with economical energy consumption [4]. The raw material was composed of 86-95% window glass waste, 4-13% sodium silicate and 1% calcium carbonate as a foaming agent. The process temperature range was 855-873 ºC and the average heating rate was between 14.5-15.5 ºC/min. The specific energy consumption had relatively low values between 1.13-1.19 kWh/kg. The main physical and mechanical characteristics of the product were: apparent density between 0.40-0.45 g/cm^3, thermal conductivity between 0.076-0.081 W/m·K and compressive strength in the range 4.9-6.2 MPa.

According to paper [5], a glass foam with very low apparent density (between 0.20-0.26 g/cm^3), low thermal conductivity (in the range 0.056-0.070 W/m·K) and high compressive strength (between 4.6-5.8 MPa) was obtained using window glass waste (83.0-83.7%), glycerol (1.0-1.8%), sodium silicate (5.3-7.5%) and water (7.7-10.0%). The weight ratio between sodium silicate and glycerol had values between 4.17-5.30. The heat treatment occurred in the temperature range of 810-824 ºC in a 0.8 kW-microwave oven of the Daily Sourcing & Research company. The specific energy consumption of the process had very low values (between 0.81-0.88 kWh/kg).

2. METHODS AND MATERIALS

2.1 Methods

In the current paper, the authors aim to manufacture porous glass foams with high mechanical strength using the nonconventional microwave heating technique, which allows to obtain low specific energy consumptions. Taking into account the results previously obtained both in the industrial production and in several small-scale experiments, including those of the authors, the solution of the mixed use of a liquid carbonic foaming agent (glycerol) and a solid foaming agent (calcium carbonate or sodium carbonate) was selected. As the raw material heating technique, the microwave heating was adopted.

The basic principle of the foaming process of glass waste with glycerol ($C_3H_8O_3$) consists in its oxidation at temperatures above 260 ºC in the oxidizing atmosphere of the oven releasing, in a first stage, carbon dioxide (CO_2) and water vapor (H_2O).

$$C_3H_8O_3 + 3.5\ O_2 = 3CO_2 + 4H_2O \quad (1)$$

The use of glycerol as a foaming agent is associated always with the use of sodium silicate (Na_2SiO_3) known also as “water glass”,that has the role of slowing down the glycerol decomposition in several compounds from CO_2 to pure carbon as well as hydroxyl compounds, by enveloping the fine particles of carbon.

The glycerol and sodium silicate, which are liquid materials, are diluted with water to reduce the viscosity of the aqueous solution and the mixture is prepared separately from the other solid components (glass waste and solid foaming agent) [6, 7].

As mentioned above, calcium carbonate ($CaCO_3$) and sodium carbonate (Na_2CO_3) were used separately as solid foaming agents in the experiments. The two types of carbonate decompose into CO_2, which is blocked in the form of bubbles in the viscous mass of the glass and calcium oxide (CaO), respectively sodium oxide (Na_2O) which are incorporated in the composition of molten glass.

$$CaCO_3 = CaO + CO_2 \quad (2)$$

$$Na_2CO_3 = Na_2O + CO_2 \quad (3)$$

$CaCO_3$ and Na_2CO_3 behave differently in terms of the volume expansion degree of the foamed material. According to [8], Na_2O incorporated in the glass composition causes a higher degree of phase separation, that leads to the formation of a glass melt with low surface tension, but higher viscosity that limits the volume expansion.

The decomposition reaction of $CaCO_3$ occurs at temperatures above 750 ºC [9]. The Na_2CO_3 decomposition research [10] has shown that the process begins after 850 ºC.

The heating technique adopted in the experiments was that of predominantly direct microwave heating. The experimental equipment was a 0.8 kW microwave oven of the type commonly used in the household for food preparation, but adapted for operation at high temperatures. The raw material mixture, pressed into a metal mold and then released, was placed freely on a metal plate placed on a bed of ceramic fiber mattresses at the base of the oven. The pressed material was protected from the microwave field (emitted by the waveguide provided in one of the side walls of the oven) by a ceramic tube made of SiC and Si_3N_4 with a wall thickness of 2.5 mm. The tube was placed on the bed of ceramic fiber mattresses and its upper opening was covered with a ceramic lid of the same material as the tube. Because the heat transfer mode characteristic of the direct microwave heating is completely different from the conventional one, the heating being initiated in the core of the material and the heat is transmitted from the inside to the peripheral areas, it was necessary the very efficient thermal protection of the material, ceramic tube and lid with ceramic fiber mattresses. Since the microwave heating is not fully direct, part of the microwave energy is absorbed in the wall of the ceramic tube, which heats up quickly and intensely due to the high microwave susceptibility of the ceramic tube components. The outer wall of the tube being thermally protected with ceramic fiber, the waves energy absorbed and converted into heat is transferred to the material subjected to heating by thermal radiation. The control of the heating process was performed with a radiation pyrometer placed above the oven at about 400 mm, the hot surface of the material being viewed through holes provided axially in the upper metal wall of the oven, ceramic lid and ceramic fiber mattresses that protect the lid. In Figure 1 images of the oven (a), ceramic tube (b), ceramic fiber thermal protection of the tube and lid (c) as well as the constructive scheme of the experimental equipment (d) are shown.

2.2 Materials

The materials used in the experiments were: colorless container glass waste as raw material, calcium carbonate, sodium carbonate, glycerol as foaming agents and sodium silicate as enveloping agent.

The colorless container glass waste had the following chemical composition: 71.7% SiO_2; 1.9% Al_2O_3; 12.0% CaO; 1.0%MgO; 13.3% Na_2O; 0.05% Cr_2O_3 and 0.05% other oxides [11]. The waste was broken, ground in a ball mill and sieved at a grain size below 80 μm.

The calcium carbonate and sodium carbonate were ground in a small laboratory device at a grain size below 40 μm.

The glycerol and sodium silicate being liquid materials were separately prepared.

a b c d

Figure 1. The experimental microwave equipment

a – 0.8 kW-microwave oven; b – SiC + Si_3N_4 ceramic tube; c – ceramic fiber thermal protection; d – constructive scheme of the experimental equipment: 1 – microwave oven; 2 – ceramic tube; 3 – ceramic lid; 4 – metal plate; 5 – pressed mixture; 6 – metal support; 7 – ceramic fiber mattress; 8 – waveguide; 9 – radiation pyrometer.

3. RESULTS AND DISCUSSION

3.1 Results

Four experimental variants were adopted for manufacturing some glass foam samples with high mechanical strength (Table 1). The first two variants were based on colorless glass waste and aqueous mixtures of glycerol and sodium silicate. Variant 3 contained colorless glass waste, an aqueous mixture of glycerol and sodium silicate and calcium carbonate as a solid foaming agent, while variant 4 contained colorless glass waste, an aqueous mixture of glycerol and sodium silicate and sodium carbonate as a solid foaming agent.

Table 1. Experimental variants

Component	Variant, wt.%			
	1	2	3	4
Colorless glass waste	83.1	82.7	83.2	78.2
Calcium carbonate	-	-	1.0	-
Sodium carbonate	-	-	-	1.0
Glycerol	1.8	1.0	1.0	2.0
Sodium silicate	7.4	12.0	3.0	7.8
Water	7.7	4.3	11.8	11.0

The main functional parameters of the sintering/foaming manufacturing process is presented in Table 2 and the physical, mechanical and microstructural characteristics of the glass foam samples are shown in Table 3.

Analyzing the data in Table 2, it can observe firstly the excellent energy efficiency of the glass foams manufacturing process due to the use of the predominantly direct microwave heating. The specific energy consumption of the process is very low (between 0.96-1.10 kWh/kg), the lowest value corresponding to sample 2 performed of glass waste (82.7%), glycerol (1.0%), sodium silicate (12.0%) and water (4.3%), while the highest value was reached by the sample 4 manufacturing of glass waste (78.2%), sodium carbonate (1.0%), glycerol (2.0%), sodium silicate (7.8%) and water (1.0%).

The sintering/foaming process temperature of the glass foam varied between 830-853 ºC. The maximum temperature of this range corresponded to variant 4, whose duration was of 46 min, the average heating rate being of 18.1 ºC/min, the lowest of the heating rate values of the for variants (18.1-20.2 ºC/min). The highest index of volume growth was 2.40 corresponding to variant 3, which used two types of foaming agent (glycerol and calcium carbonate) and the lowest value belonged to sample 4, which used glycerol and sodium carbonate.

Table 2. Main functional parameters of the sintering/foaming process

Variant	Dry/wet raw material amount g	Sintering/ foaming temperature ºC	Heating time min	Average rate, ºC/min		Index of volume growth	Glass foam amount g	Specific energy consumption kWh/kg
				Heating	Cooling			
1	536/577.3	840	41	20.0	5.9	2.20	520	0.99
2	536/559	830	40	20.3	6.2	2.10	520	0.96
3	536/599.2	835	43	19.0	6.0	2.40	518	1.04
4	536/595	853	46	18.1	5.8	2.00	521	1.10

Table 3. Physical, mechanical and microstructural characteristics of the glass foam samples

Variant	Apparent density g/cm^3	Porosity %	Thermal conductivity W/m·K	Compressive strength MPa	Water absorption %	Pore size mm
1	0.23	89.0	0.058	6.4	1.5	0.20 – 0.40
2	0.24	88.6	0.051	6.3	1.3	0.05 – 0.20
3	0.38	81.9	0.078	9.5	3.5	0.20 – 0.50
4	0.50	76.2	0.149	16.6	3.8	0.15 – 0.45

The physical, mechanical and microstructural characteristics of the glass foam samples were measured by current methods. The apparent density was measured by the gravimetric method [12] and the compressive strength was determined using a Stable Micro Systems TA XT Plus Texture Analyzer. The thermal conductivity was measured by the guarded-comparative-longitudinal heat flow (ASTM E1225-04 standard) and the porosity was calculated by the method of comparing the true and apparent density [13]. The water absorption was determined by the water immersion method (ASTM D570 standard). The samples microstructure was examined with a Smartphone Digital Microscope.

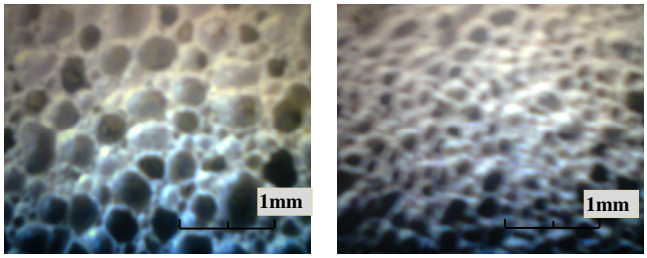
The physical, mechanical and microstructural characteristics of the glass foam samples are shown in Table 3.

Examining the data in Table 3, the low values of apparent density (between 0.23-0.50 g/cm^3) and thermal conductivity (between 0.051-0.149 W/m·K) as well as the high values of compressive strength (between 6.3-16.6 MPa) are highlighted. Pore sizes were very small (between 50-500 μm). The highest compressive strength was 16.6 MPa and corresponded to variant 4. It can be seen that the samples with high compressive strengths are those in which both aqueous solution of glycerol and sodium silicate as well as a solid foaming agent (sodium carbonate in variant 4 and calcium carbonate in variant 3) were used.

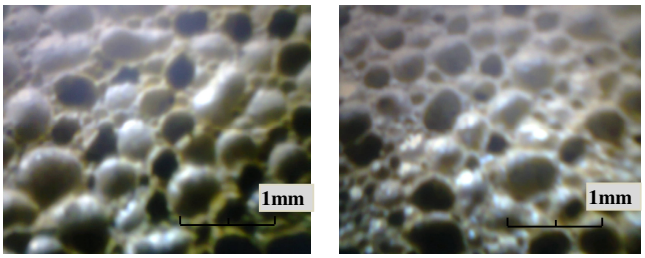
Figure 2 presents pictures of the four glass foam samples and Figure 3 shows microstructural images of the samples.

a b c d

Figure 2. Pictures of section of glass foam samples
a – sample 1; b – sample 2; c – sample 3; d – sample 4.



a b



c d

Figure 3. Microstructural images of the glass foam samples
a – sample 1; b – sample 2; c – sample 3; d – sample 4.

3.2 Discussion

Considering the objective of the paper, sample 4 was the best because it reached the highest value of the compressive strength (16.6 MPa) and simultaneously, had adequate physical and thermal characteristics to be an excellent thermal insulating material usable in construction (apparent density of 0.50 g/cm^3, porosity of 76.2% and thermal conductivity of 0.149 W/m·K). Due to the use of a liquid foaming agent, the pore size was very low (0.15-0.45 mm).

By comparison with results obtained in the world and presented in the literature [3], a similar material was manufactured by conventional heating at 850 ºC with a much lower heating rate (10 ºC/min) of 92% container glass waste, 3.5% yellow glycerol from biodiesel production (containing 80% glycerol, 10% methanol, 10% water), 3.5% sodium silicate and 1% sodium carbonate, i.e. a manufacturing recipe in which the proportions of materials differed significantly from the recipe used in the current paper. The product had higher density and thermal conductivity compared to the sample made at Daily Sourcing & Research company (0.67 g/cm^3 and 0.260 W/m·K, respectively) and a similar compressive strength (16.0 MPa).

The specific energy consumption of the experimentally manufactured sample by predominantly direct microwave heating is low (1.10 kWh/kg). The comparison with the energy consumption of the similar sample from the paper [3] could not be made because this functional parameter of the process was not shown in the paper.

4. CONCLUSION

The paper aimed to manufacture glass foams with thermal insulating characteristics and a very high compressive strength usable in construction as a replacement for existing materials.

Several manufacturing recipes were tested, the optimal recipe being composed of 78.2% container glass waste, 1.0% sodium carbonate, 2% glycerol, 7.8% sodium silicate and 11.0% water. The material

was pressed into a metal mold, then released and deposited freely between the walls of a ceramic tube covered with a lid made of SiC and Si_3N_4, a material with high microwave susceptibility. The sintering/foaming temperature was 853 C.

The thermal insulating characteristics of the product were determined by the apparent density of 0.50 g/cm^3, the porosity of 76.2% and the thermal conductivity of 0.149 W/m·K. The compressive strength had the value of 16.6 MPa.

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