GLASS FOAM FROM BOROSILICATE GLASS WASTE PRODUCED IN MICROWAVE FIELD

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ABSTRACT: The paper presents experimental results of producing glass foam from borosilicate glass waste (laboratory ware) using the microwave energy. More variants of foaming agents (silicon carbide, calcium carbonate and activated carbon) and additives (coal ash and disodium phosphate) were tested in the Romanian company Daily Sourcing & Research. The glass foams have characteristics specific to those made of borosilicate glass waste by conventional heating, i. e. relatively low apparent density (0.34 – 0.47 g/ cm^3), but higher than those resulting from soda-lime glass waste and compressive strength (1.5 – 2.5 MPa) higher than theirs. As glass foams mainly made of soda-lime glass waste, and those made of borosilicate glass waste are suitable for use as insulating material in construction.
KEYWORDS: glass foam, borosilicate glass waste, microwave, foaming agent, insulating material.

1. INTRODUCTION

Unlike the conventional manufacturing techniques of glass foams used worldwide, the production methods of these porous materials, tested in recent years by the Romanian company daily Sourcing & Research Bucharest, are based on the microwave energy. Generally, this nonconventional heating technique, fast, clean and economical, is currently applied in industrial processes on a very small scale. However, recently it was demonstrated experimentally that the microwave energy can be used efficiently in heating processes for a wide range of materials: organics, ceramics, polymers, glass, sol-gel, metals and composites [1, 2].

On the use of microwaves to heat the glass, experimental results showed a strong correlation between its electrical conductivity and the microwave absorption. It has also been found that a high concentration of alkali (Na_2O, K_2O) in the glass composition greatly favors the efficiency of heating it [1, 3].

A strong absorption of the electromagnetic wave radiation was achieved by preheating the silicate glasses or by application of microwave absorbing coating or by hydration [1, 2].

The Romanian company has already obtained significant results using microwave energy, the glass foams produced from soda-lime glass waste (packaging glass and flat glass) corresponding to the requirements of the insulating materials (light or dense) for construction and having physical, mechanical and morphological features similar to those of the glass foams manufactured by conventional techniques [3 – 6].

The latest research has been aimed at testing the production in microwave field of glass foams from borosilicate glass waste. Manufacturing this glass type has a significant spread in the world. The borosilicate glass production in the EU countries in 2009 was estimated at 3 mil. tonnes, representing about 10% from the total production of glass. Using boron in the glass chemical compositions, numerous types of borosilicate glass are manufactured: heat resistant cookware, laboratory ware, liquid crystal display screens, exterior lighting, industrial equipment. By its specific structure with glassy network, boron has an important influence on this feature [7].

Unlike the soda lime glasses, whose main components are SiO_2, Na_2O and CaO, the borosilicate glasses have in their composition mainly SiO_2 and B_2O_3. The coefficient of linear thermal expansion is much lower compared to the soda-lime glasses, the thermal shock resistance is medium-high compared to the low resistance of the soda-lime glasses and the chemical resistance is high compared to their medium resistance [8].

In the category of borosilicate glass enter: textile fiber glass, low thermal expansion borosilicate glass, fiber wool glass, alkali-free lad glass (for electrical applications), solid oxide fuel cell glass-ceramic seal etc., varying by the weight proportion of the components [9].

The literature presents experimental results obtained worldwide on the production of glass foams from borosilicate glass waste. In all cases the heating techniques used are conventional. According to [10], black carbon is recommended as foaming agent for borosilicate glasses as well as for conventional windows glasses (flat glasses). The same work considers necessary the presence of oxygen-releasing agents as SO_3 in the glass composition or Fe_2O_3 or Sb_2O_3 as additives in the powder mixture of glass and foaming agent.

Results of research on foaming borosilicate glass waste with black carbon [11] indicate an optimal heating speed of the powder mixture of 8 ºC/ min and dimensions of foaming agent particle of 150 μm to obtain a homogeneous structure of glass foam.

In the paper [12] is shown the favorable impact of a suitable addition (about 9 wt.%) of Sb_2O_3 in the composition of some basic glasses with 60% SiO_2, 13% H_3BO_3, 5% Al_2O_3, 17% Na_2CO_3 and 5% K_2CO_3 on the obtaining glass foam with low apparent density, low water absorption, high compressive strength, microstructural homogeneity. The black carbon (1 wt.%) as foaming agent and an addition of 6 wt.% disodium phosphate (Na_2HPO_4) were used. The sintering temperature was 750 – 800 ºC.

According to [13], glass foams with apparent density of 0.3 g/ cm^3 can be obtained with an addition of 0.6 wt.% Sb_2O_3. Additions of 0.2 – 0.3 wt.% Sb_2O_3 lead to the increase of porosity with 10 – 15% and of compressive strength with 20% [14].

Glass-ceramic foams with micrometric crystal of wollastonite and cristobalite from borosilicate glass waste (from washing machines), characterized by high content of CaO, were obtained at low sintering temperatures (850 – 900 ºC). The apparent density of the glass foams is 0.5 g/ cm^3, the porosity 78 – 79% and the material has closed pores [15].

The experiments conducted by Daily Sourcing & Research were focused on the low expansion borosilicate glass, being laboratory ware waste (mainly 80.6 – 81.0 wt.% SiO_2, 13.0 wt.% B_2O_3 [16]).

2. METHODS AND MATERIALS

2.1 Methods

The microwave heating process of the powder raw material for foaming was achieved in a 0.8 kW-domestic microwave oven adapted to operate at high temperature. The rotating mechanism of the material heating was dismantled, the process being in a static position. The microwave generator is placed in one of the sidewalls of the oven. The electricity consumption is counted.

The glass waste finely ground (below 150 μm) in a laboratory equipment is mixed and homogenized together with the foaming agent (silicon carbide, calcium carbonate or carbon) and the used additive (coal ash or disodium phosphate) in a small mechanical installation. The water addition is made after obtaining the powder mixture. The wetted material is loaded into a metal tube (from two semi cylindrical halves joined together) placed on a metal plate with thickness of 5 mm, deposited on a support made of rolled sheet (Fig. 1c). The support is placed on ceramic fiber mats deposited at the base of the oven.

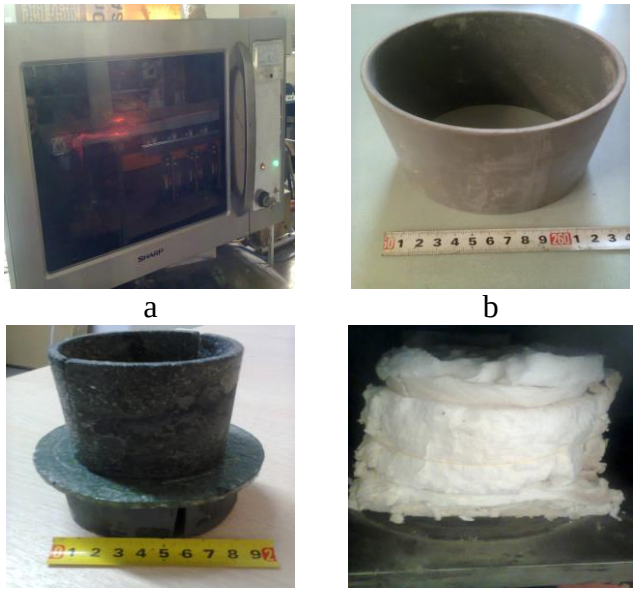

Figure 1. The microwave equipment

a – 0.8 kW-microwave oven; b – microwave susceptible tube (SiC + Si_3N_4); c – metal tube on a metal plate; d – ceramic fiber mats for thermal protection.

Concentric, a SiC + Si_3N_4 (microwave susceptible material) tube with thickness of 3.5 mm is positioned (Fig. 1b). The tube is provided with a cover of the same material. The sidewall of the tube and the upper cover surface are thermally protected with ceramic fiber mats (Fig. 1d).

The entire construction described above is introduced into the microwave oven (Fig. 1a). The control of the heating process is achieved through a PYROVAR type pyrometer (with the measuring domain between 600 – 2000 ºC) positioned above the oven, with the possibility to see the upper material layer in the metal tube through the holes in the cover made of susceptible ceramic material, the mattress above the cover and the upper wall of the oven.

The manufacturing process of glass foam is stopped when foam is found. The thermally protected

construction is kept in the oven for about 30 min., then is taken out and allowed to cool freely. After the complete cooling of material, this is released from the metal tube in which the foaming has occurred.

The glass foam samples, resulted after the sintering/ foaming experimental process, were tested in laboratory to determine the physical, mechanical and morphological characteristics. Apparent density, porosity, thermal conductivity, compressive strength, hydrolytic stability, water absorption and the crystallographic structure were determined by the current methods [17 – 20].

2.2 Materials

Between the known borosilicate glasses, low thermal expansion glass (laboratory ware) was selected for the experimentation of producing glass foam in microwave field. Its chemical composition is indicated in literature [7, 9, 21, 22] being shown in Table 1. The borosilicate glass waste was broken, ground in a small laboratory equipment and sieved at the maximum size of 150 μm.

Three foaming agent types were adopted for experiments: silicon carbide (between 63 – 80 μm), calcium carbonate (below 40 μm) and activated carbon (below 32 μm).

The coal ash (with the chemical composition shown in Table 1 and granulation below 150 μm) as well as disodium phosphate (with granulation below 150 μm) were the additives used in experiments.

Table 1. Chemical composition of borosilicate glass and coal ash, wt.%

Material	Borosilicate glass	Coal ash
SiO_2	81.0	46.5
B_2O_3	13.0	-
Al_2O_3	2.0	23.7
Na_2O	4.0	6.0
K_2O	-	4.1
CaO	-	7.9
MgO	-	3.2
Fel_2O_3	-	8.6

3. RESULTS AND DISCUSSION

3.1 Materials

The three variants adopted for producing glass foam from borosilicate glass waste, tested in an adapted 0.8 kW-domestic microwave oven, consist in the use of three different foaming agents and suitable additives, whose weight proportions are shown in Table 2.

Table 2. Raw material components used in experiments, wt.%

Raw material	Variant 1	Variant 2	Variant 3
Borosilicate glass waste	87.9	98.7	92.8
Silicon carbide	3.0	-	-
Calcium carbonate	-	1.3	-
Activated carbon	-	-	1.0
Coal ash	9.1	-	-
Disodium phosphate	-	-	6.2
Water addition	15.0	10.0	10.0

The main functional parameters of the sintering/ foaming process, corresponding to the three experimental variants are presented in Table 3.

Table 3. Functional parameters of the sintering/ foaming process

Parameter	Variant 1	Variant 2	Variant 3
Raw material quantity -dry (g) -wet (g)	 256.4 294.7	 264.8 292.3	 250.6 275.7
Heating duration (min)	75	50	52
Sintering/ foaming temperature (°C)	970	830	820
Average speed (°C/min) -heating -cooling	 12.6 6.5	 16.1 6.2	 15.3 6.4
Index of volume growth	1.85	1.80	1.90
Glass foam quantity (g)	248.0	256.0	243.0
Specific consumption of electricity (kWh/ kg)	4.03	2.62	2.84

The physical, mechanical and morphological features of the three experimental samples are shown in Table 4.

Table 4. Physical, mechanical and morphological features of glass foam samples

Feature	Variant 1	Variant 2	Variant 3
Apparent density (g/cm^3)	0.38	0.47	0.34
Porosity (%)	82.7	78.6	84.5
Compressive strength (MPa)	1.5	2.4	2.5
Thermal conductivity ($W/m \cdot K$)	0.054	0.078	0.055
Water absorption (%)	2.7	5.7	2.1
Pore size (mm)	0.8 – 2.0	0.8 – 1.3	1.0 – 2.5

Images of the glass foams (in view and longitudinal section respectively) are shown in Fig. 2.

Sample 1 Sample 2 Sample 3

Figure 2. Images of the glass foams

The tests for determining the hydrolytic stability of samples, using 0.15 ml of 0.01M HCl solution to neutralize the extracted Na_2O, showed that the stability joins in the hydrolytic class 2, the extracted Na_2O equivalent being in the range 34 - 52 μg.

The XRD analysis to identify the crystalline phases from the structure of glass-ceramic foams were carried out on the sample 1 and 3, using a X-ray diffractometer Bruker-AXS Advance with CuKα radiation. For the sample 1 the main crystalline phase identified was wollastonite and traces of cristobalite. For the sample 3 the crystalline phases identified was sodium aluminium phosphate and cristobalite. The sample 2 has not crystalline phases.

3.2 Discussion

The manufacturing methods of glass foam from borosilicate glass waste tested experimentally are based on the two main type of foaming agents: neutralizers and redox. The agents from the first category ($CaCO_3$, $CaMg(CO_3)_2$) decompose releasing the gaseous product (CO_2), while the agents from the second category (C, SiC, Si_3N_4) reacts with the glass forming gaseous products (CO_2, CO, N_2) depending on the composition of the used foaming agent [23].

The borosilicate glasses as well as the flat glasses, are efficiently foamed using SiC or Si_3N_4 [23].

Starting from these specifications, the three variants of producing glass foam described above were adopted. Comparing the three variants of producing glass foam, based on the data from Tables 3 and 4, the most advantageous in terms of quality and energy consumption is the variant 3, which uses actived carbon as foaming agent and Na_2HPO_4 as fluidizing additive. The sintering/ foaming process temperature is 820 ºC, the process duration 52 min and the heating speed 15.3 ºC/ min. The volume growth of the foamed material is 90%. The glass foam has apparent density 0.34 g/ cm^3, thermal conductivity 0.055 W/ m · K and compressive strength 2.5 MPa. The product is very poorly water-absorbing (2.1%) and the sample microstructure in transversal section is homogeneous, with pore size between 1.0 – 2.5 mm.

The first variant, using SiC (3%) and coal ash (9.1%), allows obtaining homogeneous microstructure (pore size 0.8 – 2.0 mm uniformly distributed), but the process temperature (influenced by the presence of coal ash in the mixture composition) reaches 970 ºC, increasing the energy consumption. Also, the compressive strength is the lowest between the three variants, its value (1.5 MPa) being high enough for the insulating material requirements.

The variant 2, using $CaCO_3$ as foaming agent, requires a sintering/ foaming temperature relative low (830 ºC) and the shortest process duration (50 min). However, the product has the highest values of apparent density (0.47 g/ cm^3) and thermal conductivity (0.078 W/ m · K), but also a high compressive strength (2.4 MPa). The sample microstructure is homogeneous, with small pores (0.8 – 1.3 mm). However, the water absorption has the highest value (5.7%) compared to the other two variants.

It should be mentioned that all the experimentally performed samples (in microwave field) are suitable for use in construction as insulating material and their physical, mechanical and morphological features are similar to those manufactured by conventional heating methods.

4. CONCLUSION

The research conducted by the Romanian Company Daily Sourcing & Research aimed to obtain in microwave field glass foams suitable for construction as insulating material from borosilicate glass waste.

Three foaming technique of glass waste were tested in an adapted 0.8 kW-domestic microwave oven, using silicon carbide and coal ash (variant 1), calcium carbonate (variant 2) and activated carbon and disodium phosphate (variant 3).

All the experimentally performed samples (in microwave field) are suitable for use in construction as insulating material and their physical, mechanical and morphological features are similar to those manufactured by conventional heating methods.

The most advantageous in terms of quality and energy consumption is the variant 3. The sintering/ foaming process temperature is 820 ºC and the process duration is 52 min. The glass foam has apparent density 0.34 g/ cm^3, thermal conductivity 0.055 W/ m · K and compressive strength 2.5 MPa. The product is very poorly water-absorbing (2.1%)

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