FOAM GLASS GRAVEL MADE OF RECYCLED GLASS WASTE AND SILICON CARBIDE BY MICROWAVE HEATING

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Abstract: The paper presents recent achievements in the microwave use for manufacturing foam glass gravel from recycled glass waste and silicon carbide. The aim was to obtain a product with physical and mechanical characteristics almost similar to those of industrially manufactured materials by conventional heating techniques, but with a higher energy efficiency. A foam glass with the thermal conductivity of 0.075 W/m·K and the compressive strength of 7.5 MPa was experimentally obtained. The specific energy consumption was of 1.0 kWh/kg comparable with the industrial processes and it could reach values up to 25% lower by using a high power industrial microwave equipment.

Keywords: foam glass gravel, glass waste, silicon carbide, microwave, compressive strength, thermal conductivity, energy efficiency

1. INTRODUCTION

The foam glass gravel has several unique physical and mechanical characteristics of which the correlation between the thermal conductivity and the compressive strength is the main asset of this material type. According to [1], the thermal conductivity of the best cellular glasses is approximately twice as high compared to loose fill thermal insulations existing on the market for similar compressive strength. Other peculiarities of the foam glass gravel (low apparent density, very low water permeability, fine porosity, resistance to fire, rodents, bacteria, no toxicity, chemical stability, etc. [2]) contribute to the remarkable quality of this product obtained from recycled glass waste. The application field includes insulation fill in building foundation, lightweight fill material for landscaping, roof gardens, green roofs, new floor in old buildings, road and railway construction, bridge abutments and retaining walls, insulating of underground pipelines and storage tanks, etc. [3].

The manufacturing technique of the product does not involve special raw materials and processing conditions. Different glass waste types (container glass of various colors or flat glass) can be used without special cleaning conditions. Generally, two foaming agent types are used: liquid (glycerol) and solid (silicon carbide, soot or coal powder, calcium carbonate, gypsum, sodium silicate, manganese oxide) [4, 5]. The liquid foaming agent allows to obtain a very fine porosity, unlike the solid agent [6]. The manufacturing recipes of the foam glass gravel producers (Geocell Schaumglas, Misapor, Glapor, Glamaco, Geoglass, Refaglass) include one or more foaming agents mentioned above [1]. The industrial facilities for the production of foam glass gravel consist of tunnel ovens with metal conveyor belt powered by conventional energy sources (fossil fuels or electrical resistances).

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The foamed product has high compressive strength (5-6 MPa or more) and low thermal conductivity (0.06-0.09 W/m·K) [4, 5, 7].

The literature provides more information on the manufacture of foam glass gravel using glycerol as a liquid foaming agent. Data on manufacturing recipes of the main industrial manufacturers of this foam glass gravel type (Glapor and Glamaco), functional parameters and characteristics of their products are presented in the literature [5, 8, 9]. Thus, using 87% glass waste, 1% glycerol and 12% sodium silicate, Glapor obtained a product with bulk density of 0.13-0.21 g/cm^3 and compressive strength of 4.9-6 MPa. Glamaco used 95% glass waste, glycerol, calcium carbonate and sodium silicate accounting for 5% and obtained a foam glass gravel with bulk density of 0.15-0.20 g/cm^3, thermal conductivity of 0.06-0.08 W/m·K and compressive strength of 4-6 MPa. In our previous studies we carried out research on the manufacture of foam glass gravel with glycerol and sodium silicate by the unconventional method of using microwaves obtaining products with characteristics similar those industrially manufactured (thermal conductivity between 0.057-0.063 W/m·K, compressive strength up to 5.9 MPa, homogeneous porous structure with pore size below 0.9 mm and specific energy consumption between 0.83-0.88 kWh/kg. The results are being published in a Romanian journal. Referring to the industrial manufacture of foam glass gravel using solid foaming agents the information provided by the literature is much more limited. Our team has experimentally obtained in microwave field a foam glass gravel made of glass waste (90.5-91.7%), calcium carbonate (3-5%) and borax (4.5-5.3%) with the following characteristics: thermal conductivity between 0.087-0.110 W/m·K, compressive strength up to 6.3 MPa and pore size from 0.7-0.9 mm up to 1.0-1.7 mm [2]. Of course, there are several works that present conventional techniques for the production of glass-ceramics from glass waste, coal ash (between 10-20%) and silicon carbide (between 2-5%) leading to obtaining products with thermal insulation characteristics in the form of plates or blocks having moderate mechanical strength [10, 11]. Such products were experimentally obtained too by the microwave irradiation in the company Daily Sourcing & Research [12, 13] having characteristics similar to those manufactured by conventional techniques. However, it should be mentioned that there is a fundamental difference between these products and the foam glass gravels and therefore is not the subject of this paper.

A fast, economical and ecological unconventional heating technique known since the mid-20th century, but industrially used to a very small extent, is the microwave heating. According to [14], only in the last 10-15 years it has been found that the microwave irradiation is suitable for several material types such as organics, ceramics, metals, polymers, glass, etc., but the industrial application of the technique is still in an experimental stage. In the last three years, the Romanian company Daily Sourcing & Research Bucharest obtained important results in the research of glass foam making in microwave field, published in several Romanian and international journals [12, 13, 15-18].

The current paper refers to tests performed in the field of manufacturing the foam glass gravel using a solid foaming agent (silicon carbide), aiming the influence of the functional parameters that determine the energy efficiency of the process (temperature, duration, heating and cooling rates) on the physical, mechanical and morphological characteristics of the foamed product due to the application of the unconventional heating technique, that constitutes the originality of the study.

2. MATERIALS AND METHODS

2.1. Materials

The materials that composed the load for the experimental manufacture of foam glass gravel were container glass waste of different colors commonly available on the market (colorless, green and amber) in a weight ratio of 50/20/30 considered close to reality taking into account the average quantities of recycled glass waste [18] as well as silicon carbide as a foaming agent. Knowing the previously determined chemical composition [18] of the three glass waste types shown in Table 1, an average composition of the waste used in experiments was calculated.

The glass waste was thermally washed at 250 ºC to remove the organic contaminants, ground in a ball mill and sieved at a granulation below 300 μm.

The silicon carbide was used at the grain size below 40 μm without other processing techniques as purchased from the market.

Table 1. Chemical composition of the glass waste.

Glass waste type	Chemical composition (wt.%)									
	SiO_2	Al_2O_3	CaO	Fe_2O_3	MgO	Na_2O	K_2O	Cr_2O_3	SO_3	Other oxides
Colorless	71.7	1.9	12.0	-	1.0	13.3	-	0.05	-	0.05
Green	71.8	1.9	11.8	-	1.2	13.1	0.1	0.09	-	0.01
Amber	71.1	2.0	12.1	0.2	1.1	13.3	0.1	-	0.05	0.05
Average	71.54	1.93	11.99	0.06	1.07	13.26	0.05	0.043	0.015	0.042

2.2. Methods

Among the foaming agents used in the glass foam manufacturing process, the silicon carbide is recognized for its high efficiency. The weight ratio of silicon carbide in the mixture with the glass powder can be between 2-5% [10, 11], but it was found that the use of only 2% is sufficient for a proper foaming. In addition, it allows reducing the porosity and increasing the mechanical strength of the product [19]. The silicon carbide causes the glass foaming by producing gaseous bubbles as a result of oxidation reactions shown below.

$$SiC + 2\ O_2 = SiO_2 + CO_2 \quad (1)$$

$$SiC + O_2 = SiO + CO \quad (2)$$

The DTA/TGA analysis of the pure silicon carbide and the starting mixture indicates that the above reactions are mainly active at about 900 ºC [20, 21].

It has been experimentally found since the first tests [22] that the glass waste cannot be sintered and foamed properly by direct microwave heating due to the destructive effect on the internal structure of the material caused by the excessively high heating rate. For this reason, a ceramic crucible or tube from a microwave susceptible material was used, being placed between the microwave generating source and the glass-based material subjected to the thermal process. This technical solution allowed the partial absorption of the electromagnetic waves in the mass of the crucible or tube, which heats up rapidly and intensely and transfers the heat through thermal radiation, while a moderate microwaves proportion penetrating the ceramic wall comes in direct contact with the material, which heats it rapidly from its core the heat being then transferred to its peripheral areas [22]. This heating mode partially direct and partially indirect has proved to be suitable for any type of glass waste. The thickness of the crucible or tube wall (between 3.5-5 mm) and the nature of the used ceramic material (a composition based on silicon carbide) were the variable elements of determining an optimal ratio between the two components of the microwave heating. All previous own experiments were performed using the mixed microwave heating system and the quality of the glass foams was adequate.

The experimental equipment was a 0.8 kW microwave oven (a1) of the type used in the household adapted for high temperature operation (up to 1200 ºC). The pressed mixture (a5) consisting of ground glass waste and silicon carbide was deposited freely on a metal plate (a4) placed through a metal support (a6) on a bed of ceramic fiber mattresses (a7) at the base of the oven. A ceramic tube (a2) made of predominantly silicon carbide with the wall thickness of 5 mm was placed around the pressed mixture and provided with a lid (a3) of the same material. The outer surface of the tube and lid has been protected with ceramic fiber mattresses (a7) to avoid the heat loss outside the system. A radiation pyrometer (a9) was mounted above the oven on a central axis to visualize the surface temperature of the heated material through the holes provided in the upper metal wall of the oven, the ceramic tube lid and the ceramic fiber that protects the lid. The control of the surface temperature of the pressed material allowed to identify the end of the foaming process. Stopping the tendency to increase the temperature and starting a slight decrease is the indication of foaming in the final phase and the possibility of stopping the electricity supply of the oven. This method of controlling the sintering/ foaming process was applied during the experiments for the first variant. The following tests corresponding to variants 2-4 successively had shorter heating times compared to the duration of variant 1 and were arbitrarily adopted considering the appearance of each previous sample. The constructive scheme of the experimental equipment is shown in Figure 1a and the detail of the pressed mixture positioning on the metal plate is presented in Figure 1b.

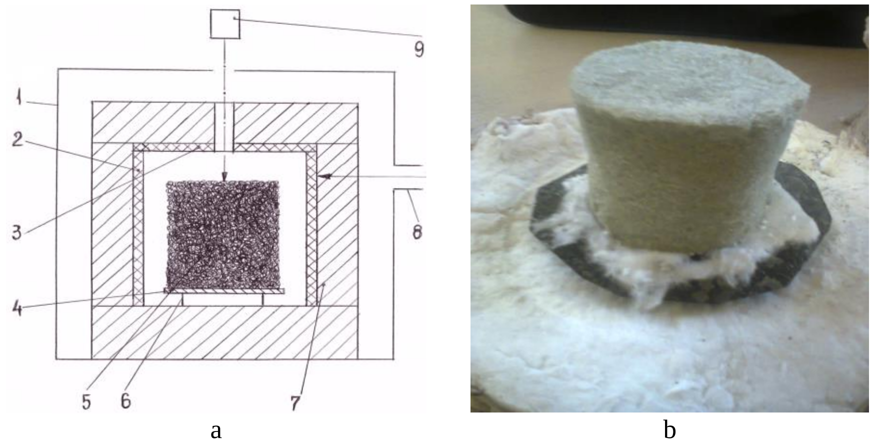


Fig. 1. The experimental equipment: a – constructive scheme; 1 – microwave oven; 2 – ceramic tube; 3 – ceramic lid; 4 – metal plate; 5 – pressed mixture; 6 – metal support; 7 – ceramic fiber; 8 – microwave source; 9 – pyrometer; b – the detail of the pressed mixture positioning.

2.3. Characterization of the foamed product

The foam glass gravel samples manufactured by the sintering/foaming technique using silicon carbide as a foaming agent were analyzed to identify their main characteristics. The bulk density was measured by the method of determining the mass of the undisturbed solid samples fully loaded in a container of known volume [23]. The dimensions of the foam glass gravel samples were below 50 mm. The determination of the porosity required the comparison between the density of the cellular product and the density of the melted and compactly cooled material [24]. The determination of the thermal conductivity was performed by measuring the thermal flow which crosses through the sample mass with thickness of 50 mm placed between two metal plates, one heated and thermally protected and the other cooled [25]. The water permeability of the porous product was measured by the water immersion method (ASTM D 570). An own conception device was used to measure the compressive strength of the samples. It developed an axial pressing force of maximum 20 tons-force with a hydraulically operated piston. The dimensions of the cylindrical samples were 80 mm for the diameter and 70 mm for the height. The compressive strength was measured up to the crack of the sample subjected to the axial pressing. The microstructural configuration of the samples was performed with a Smartphone digital microscope.

3. RESULTS AND DISCUSSION

3.1. Results

Previous own tests confirmed that the weight ratio of silicon carbide in the glass waste mixture can be reduced up to 2%, value that can be considered optimal to make a proper foam glass gravel. The experiments presented in the paper refer to the influence of the process duration and implicitly, its final temperature on the physical, mechanical and morphological characteristics of the foamed product.

The manufacturing recipe strikethrough used in experiments included 98 wt.% container glass waste, 2 wt.% silicon carbide and additionally 15.4 wt.% added water.

Four experimental variants were adopted, where the final temperature of the heating process and the heating time in variant 1 were experimentally determined according to the method presented in the chapter 2.2. For variants 2-4 the adopted heating durations were successively 50, 47 and 43 min in this order, the temperature values being measured. Experiments were carried out in duplicate to confirm the reproducibility of the foam glass gravel samples.

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

Analyzing the data in Table 2, the energy efficiency of the microwave heating process is remarkable. The heating rate is within the optimal range for obtaining a proper foaming of the glass waste (16.6-20.9 ºC/min), being determined by the process duration (between 43-55 min). Under these conditions, the specific energy consumption has values between 0.93-1.18 kWh/kg. Although the literature does not explicitly present the value of the specific energy consumption of the industrial manufacturing process of foam glass gravel, it can be deduced that the average value is around 1 kWh/kg [26], i.e. approximately equal to that achieved under experimental conditions on the very low power oven (0.8 kW).

Table 2. The main functional parameters of the sintering/foaming process.

Variant	Dry raw material/ foam glass gravel quantity (g)	Sintering/ foaming temperature (ºC)	Heating time (min)	Average rate (ºC/ min)		Index of volume growth	Specific energy consumption (kWh/kg)
				Heating	Cooling		
1	560/ 543	929	55	16.6	5.5	2.36	1.18
2	560/ 542	925	50	18.1	5.1	1.97	1.08
3	560/ 545	922	47	19.2	6.0	1.83	1.00
4	560/ 540	916	43	20.9	7.3	1.60	0.93

Table 3. Physical, mechanical and morphological characteristics of the samples.

Variant	Bulk density (g/cm^3)	Porosity (%)	Thermal conductivity (W/m·K)	Compressive strength (MPa)	Water permeability (%)	Pore size (mm)
1	0.23	83.6	0.063	5.3	0.8	0.9 – 1.9
2	0.26	81.8	0.073	7.2	0.7	0.8 – 1.4
3	0.27	80.9	0.075	7.5	0.4	0.5 – 1.0
4	0.31	77.7	0.089	8.0	0.1	0.4 – 0.7

According to [14], using a microwave equipment of high power (industrial type) the energy efficiency of the process could increase by up to 25%, that would prove the superiority of heating by the unconventional method. By associating the data in Table 3 with those in Table 2, significant variations of the physical, mechanical, and morphological characteristics are observed depending on the functional parameters of the sintering/foaming process. Thus, the bulk density varied between 0.23-0.31 g/cm^3, the thermal conductivity had values between 0.063-0.089 W/m·K, the compressive strength increased from 5.3 up to 8.0 MPa and the pore size in the material structure decreased from 0.9-1.9 mm up to 0.4-0.7 mm.

Pictures of the four foam glass gravel samples are shown in Figure 2.

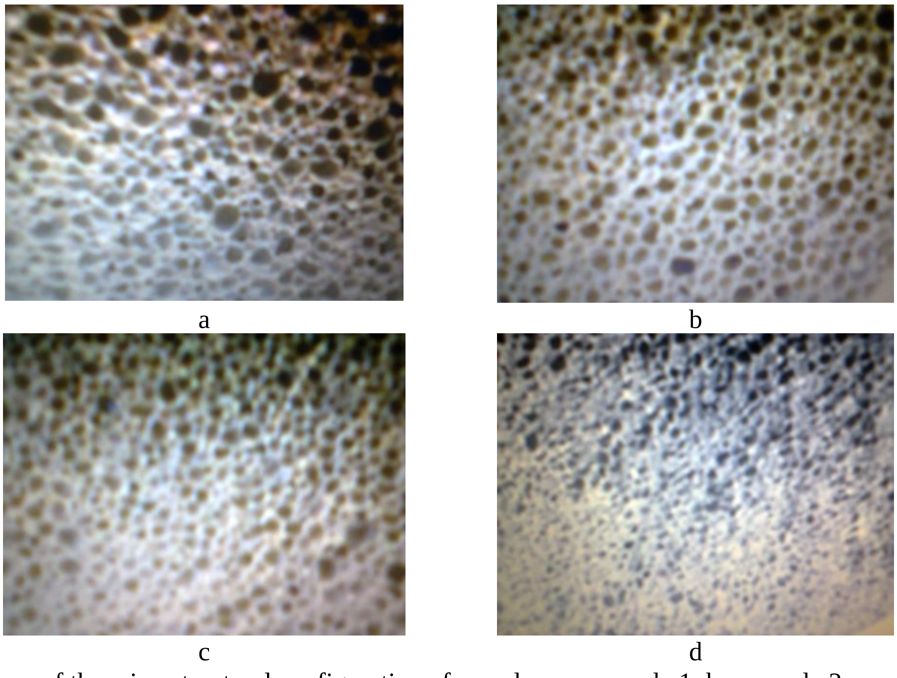
Examining the characteristics of the samples in Table 3 as well as the images in Figure 2 led to the identification of the sample considered optimal for the manufacture of foam glass gravel with silicon carbide as a foaming agent by microwave irradiation. This is the sample corresponding to the variant 3 obtained by thermal treatment at 922 ºC for 47 min, with the heating rate of 19.2 ºC/min. The specific energy consumption was 1.0 kWh/kg. The product had bulk density of 0.27 g/cm^3, porosity of 80.9%, thermal conductivity of 0.075 W/m·K, compressive strength of 7.5 MPa, water permeability of 0.4% and pore size between 0.5-1.0 mm.

Pictures of the microstructural configuration of the four samples of foam glass gravel are shown in Figure 3. The pore sizes for the four variants are indicated in Table 3. According to the pictures, the uniform pore distribution is a common feature of all samples.

a b

c d

Fig. 2. Pictures of the foam glass gravel samples: a sample 1; b – sample 2; c – sample 3; d – sample 4.



a b

c d

Fig. 3. Pictures of the microstructural configuration of samples: a – sample 1; b – sample 2;c – sample 3; d – sample 4.

3.2. Discussion

In principle, the direct microwave heating of a powder material has a high energy efficiency. Previous tests carried out in the company Daily Sourcing & Research with aluminosilicate waste (clay brick, building concrete, cement mortar, coal ash) have confirmed that the sintering/foaming process can be performed by direct microwave heating, the product having a homogeneous porous structure [27]. This heat treatment is not suitable for silicate waste, which also includes the glass. The material suffers an aggressive thermal attack from the microwave field in the core of its mass, its internal structure being largely destroyed. Several tests performed, even in the case of a glass-aluminosilicate mixture, have confirmed this effect due to the excessively high heating rate.

The adopted technical solution was to reduce the effect of the direct microwave irradiation of the material by placing a screen (ceramic crucible or tube) from a high microwave susceptible material between the microwave generating source and the material subjected to heating. A thickness of this screen of 3.5-5.0 mm has proved to be ideal to ensure its partial penetration by the electromagnetic waves, the rest being absorbed in the screen mass. Thus, a mixed heating of the material is made, both directly in its core, the heat being transferred to the peripheral areas and indirectly by the thermal radiation of the intensely heated screen towards the material.

4. CONCLUSIONS

A foam glass gravel was experimentally produced using container glass waste as raw material and silicon carbide as a foaming agent by an unconventional heating technique at high temperature (916-929 ºC).

Unlike the commonly conventional heating techniques, the microwave heating was adopted aiming to produce a foam glass gravel with similar characteristics to those industrially manufactured and high energy efficiency.

The manufacturing recipe was kept unchanged for the four experimental variants, i.e. 98 wt.% container glass waste, 2 wt.% silicon carbide and 15.4 wt.% water addition. The variants were identified only by the final temperature of the process, which had successively the following values: 929, 925, 922 and 916 ºC.

The experimental results revealed significant changes in the physical, mechanical and morphological characteristics of the samples depending on the adopted heat treatment. Thus, by decreasing the temperature from 929 to 916 ºC porosity decreased from 83.6 to 77.7%, thermal conductivity increased from 0.063 to 0.089 W/m·K and compressive strength increased from 5.3 up to 8.0 MPa. Water permeability was negligible and the pore size decreased from 0.9-1.9 mm to 0.4-0.7 mm.

Generally, the specific energy consumption had low values (around 1 kWh/kg) comparable to those industrially achieved. The process temperature influenced the consumption value, which was reduced from 1.18 to 0.93 kWh/kg as the temperature decreased.

Among the foam glass gravel samples experimentally made the most suitable for the proposed aim was considered the sample corresponding to variant 3. Heated to 922 ºC, with a specific energy consumption of 1.0 kWh/kg, the foam glass gravel sample had bulk density of 0.27 g/cm^3, porosity of 80.9%, thermal conductivity of 0.075 W/m·K and compressive strength of 7.5 MPa. The pore size was between 0.5-1.0 mm. The sample met the requirement for use as a foam glass gravel in construction.

According to the literature, using a microwave equipment of high power (industrial type) the energy efficiency of the manufacturing process of foam glass gravel could increase by up to 25%, proving the superiority of heating by the unconventional method.

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