

HIGH MECHANICAL STRENGTH CELLULAR PRODUCT AS A CONSTRUCTION MATERIAL MANUFACTURED IN MICROWAVE FIELD

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ABSTRACT: The paper presents the results of the research for the production by a nonconventional method (using microwave irradiation) of a cellular glass with high mechanical strength from flat glass waste (86 – 95%), calcium carbonate (1%) as a foaming agent and an addition of sodium silicate also called “water glass” (4 – 13%). The cellular glass samples had the apparent density between 0.40 – 0.45 g/cm³, the thermal conductivity in the range 0.076 – 0.081 W/m·K and the compressive strength between 4.9 – 6.2 MPa, having the characteristics of a foam glass gravel industrially manufactured by conventional methods and used in construction as materials resistant to high mechanical loads. The morphological characteristic specific for the thermal insulating materials, i.e. a homogeneous distribution of the pores, have been fulfilled, even in case of using a raw material (flat glass waste) which is, generally, avoided because it causes structural inhomogeneity of the glass foam. The energy efficiency of the making process is remarkable, the specific energy consumption having low values between 1.13 – 1.19 kWh/kg.

KEYWORDS: cellular glass, microwave, high mechanical strength, energy efficiency, structural homogeneity.

1. INTRODUCTION

Since the end of the 20th century, recycling the material waste (plastics, metals, glasses, etc.) has become a major concern of the civilized world, both for reasons of environmental protection and economical by replacing expensive raw materials with wastes processed at low prices to ensure similar characteristics to those of the initial materials. Generally, the glass waste is recycled in a "closed circuit" being used as raw material in the industrial manufacture processes of the new glass. This solution involves high costs. For this reason, the tendency shown worldwide in recent decades is the use of glass waste as a building material after high temperature heat treatment together with a suitable foaming agent [1]. The product obtained in the cellular form combines the physical and mechanical characteristics of the glass with the insulating properties of the foam, being able to replace materials existing on the market usable both as lightweight insulating materials for the building walls and as high hardness insulating materials for foundations, drainages, pavements, road construction, aggregate for lightweight concrete, etc. [2]. In terms of energy, the production of the cellular glass from recycled glass requires about 500 kJ/kg, while for the manufacture of the new glass about 4500 kJ/kg [3].

One of the cellular products with high mechanical strength is the foam glass gravel, made from

recycled glass waste in the form of 20 - 80 mm pieces. The industrial production of this assortment of glass foam began in the last decades of the last century in several western European countries (Switzerland, Austria and Germany) [4, 5]. The major manufacturers of foam glass gravel are Misapor Switzerland with branches in Germany, France and Austria, Geocell Schaumglas with plants in Austria and Germany, Glapor Werk Mitterteich and Glamaco, both from Germany. The manufacturing recipes are based on very high mass ratios of glass waste of various kinds, the foaming agent differing depending on the nature of the raw material. Misapor uses calcium sulfate, calcium carbonate or silicon carbide as a foaming agent, the process temperature being between 700 - 900 °C. The obtained foam glass gravel has the bulk density between 0.13 - 0.21 g/cm³ and the compressive strength between 4.9 - 6.0 MPa [6]. Geocell Schaumglas uses colored container glass waste and colorless flat glass waste in the mass ratio 9: 1. The bulk density has an average value of 0.15 g/cm³, the thermal conductivity around 0.080 W/m·K and the compressive strength of over 5 MPa [7]. Glapor commonly uses recycled glass waste (87%), sodium silicate, also known as "water glass" (12%), glycerin (1%) and minor mass ratios of kaolin. Glapor products have the bulk density 0.11 - 0.14 g/cm³, the compressive strength of over 5 MPa and the thermal conductivity between 0.052 - 0.058 W/m·K [8]. Glamaco company uses recycled glass waste

(95%), water glass, glycerol (glycerin), calcium carbonate, water and kaolin. The glass waste comes from its own glass melting facilities or from the recycling of flat glass or container glass. The foam glass gravel has the density between 0.15 - 0.20 g/cm³, the thermal conductivity between 0.060 - 0.080 W/m·K and the compressive strength in the range 4 - 6 MPa [9].

All the cellular products with high mechanical strength mentioned above are exclusively manufactured by conventional heating methods (burning fossil fuels or using electrical resistances). A much more economical and faster energy source is the microwave energy. Known since the middle of the last century, the use of microwaves as an energy source has only materialized in the household for the food preparation and in very few other industrial processes. Only in the last decade several application fields of this source have been found (organics, ceramics, metals, glass, polymers, composites, etc.) [10], but its industrial use is delayed in making itself felt.

The Romanian company Daily Sourcing & Research Bucharest has initiated in the last three years a program of experimental manufacture of different types of glass foam using the microwave energy. The experiments aimed to manufacture cellular products with characteristics almost similar to those of the products obtained by conventional techniques. Lately, there have been concerns for achieving cellular products with high mechanical strength using the same nonconventional method specified above [11-13].

1. METHODS AND MATERIALS

1.1 Methods

For producing a cellular material with high mechanical strength, a manufacturing recipe consisting of colorless flat glass waste, calcium carbonate and an aqueous solution of sodium silicate ("water glass"), was adopted. The solid materials previously processed were mixed and homogenized and then, after adding the water glass solution as a binder, were pressed at about 15 MPa in a metal mold. The pressed product was released from the mold and placed freely on a stainless plate placed on a bed of ceramic fiber mattresses at the bottom of the microwave oven. A ceramic tube made of a microwave susceptible material (silicon carbide and silicon nitride in a 4/1 ratio) with the outside diameter of 125 mm, the height of 100 mm and the wall thickness of 3.5 mm was placed on the ceramic fiber bed enclosing the pressed sample. The dimension of the wall thickness of the tube was adopted as a result of previous tests regarding the

mode of microwave penetration and absorption through this wall, so that the ratio between the penetrating waves (which come in direct contact with the powder mixture) and those absorbed (which indirectly transfer the heat by thermal radiation) to be optimal for a rapid heating and without affecting the internal structure of the foamed product. A lid of the same ceramic material was placed on the tube. The thermal protection of the tube and the material inside it was obtained with ceramic fiber mattresses. The entire construction described above including the powder mixture is introduced in a 0.8 kW-domestic microwave oven adapted for high temperature operation (up to 1200 °C). The oven has a single microwave generator mounted on one of the side walls. The control of the thermal process is carried out with a radiation pyrometer placed above the oven in its central axis, which sees the surface of the material through holes Ø 30 mm provided in the ceramic lid of the tube, the ceramic fiber mattress above the lid and the upper metal wall of the oven (Figure 1).

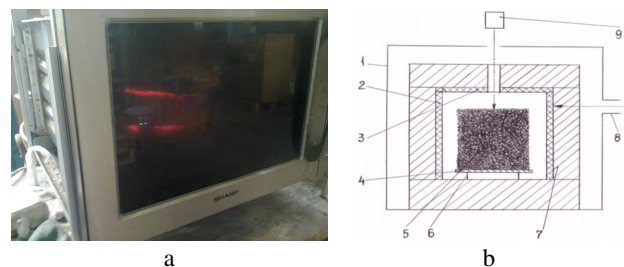


Figure 1. The experimental microwave equipment
a – 0.8 kW-microwave oven; b – constructive scheme:
1- microwave oven; 2 - ceramic tube; 3 - ceramic lid;
4 - stainless plate; 5 – pressed powder mixture;
6 - metal support; 7 – ceramic fiber protection;
8 - waveguide; 9 – pyrometer.

The microwaves, which are electromagnetic waves with frequencies between 300 MHz and 300 GHz, do not represent a direct source of energy. The condition for a material to be microwave heated is that it is microwave susceptible and is called dielectric. The interaction of dielectric materials with the electromagnetic radiation into a microwave oven has as result the energy absorption [14]. The dielectric materials absorb the electromagnetic energy in the whole volume and convert it into heat. This heating mode differs significantly from the conventional methods. Because the material itself generates heat, the heating process is volumetric and can be very fast [15]. The microwave heating eliminates the supplementary energy consumption for heating the massive components of the oven. Thus, the heating rate of the material increases and the energy consumption is reduced.

The methodology adopted for the experimentation includes four tests for the manufacture of the cellular glass with high mechanical strength, in which the mass ratio of calcium carbonate is kept constant at the value of 1%, while the proportion of water glass increases from 4% (variant 1) up to 13% (variant 4). Implicitly, the ratio of glass waste decreases from 95% (variant 1) to 86% (variant 4). Table 1 shows the composition of the four adopted variants.

Table 1. Composition of the used materials

Variant	Colorless flat glass waste wt. %	Calcium carbonate wt. %	Water glass wt. %
1	95	1	4
2	92	1	7
3	89	1	10
4	86	1	13

2.2 Materials

The materials used during the experiments were colorless flat glass waste, calcium carbonate and sodium silicate (water glass). The glass waste was mechanically crushed and then, ground in a laboratory electrical device to the grain size below 250 μm . The chemical composition of the colorless flat glass includes 71.1% SiO_2 , 1.3% Al_2O_3 , 3.9% MgO , 9.3% CaO , 14.2% $\text{Na}_2\text{O} + \text{K}_2\text{O}$ and 0.2% Fe_2O_3 [16]. Calcium carbonate was used as purchased from the market (below 40 μm). According to [17], water glass is a liquid solution that introduced into the glass waste powder homogenizes the chemical composition of the material. Due to the chemical reaction between the water glass and the surface of the glass waste particles, silicates are formed which contain chemically bound water. It is released at 600-620 $^\circ\text{C}$ and facilitates the formation of the cellular product. According to [18], water glass can be used as a

foaming agent without the need of other known agents.

2.3 Characterization of the samples

The physical, mechanical and morphological features of the foam glass samples produced in microwave field were determined in the laboratory using classic methods of analysis. The apparent density was measured by the gravimetric method [19] and the porosity was determined by the method of comparing the density of the compact material (after melting and cooling) and the densities of the porous material [20]. The compressive strength was identified by tests carried out on a hydraulically operated uniaxial press for ceramics. Using ASTM E 1225-04 standard test method for thermal conductivity of solids by means of the guarded-comparative-longitudinal heat flow technique, the thermal conductivity of the samples was measured. The hydrolytic stability of the porous material was determined using the standard procedure ISO 719: 1985 and the water absorption was measured by the usual method of the sample immersion in water.

3. RESULTS AND DISCUSSION

3.1 Results

The main functional parameters of the sintering/foaming process for manufacture in microwave field a high mechanical strength cellular product are presented in Table 2 and the physical, mechanical and morphological characteristics of the samples are shown in Table 3.

Table 2. Parameters of the sintering/ foaming process

Variant	Raw material/ glass foam amount g	Sintering/ foaming temperature $^\circ\text{C}$	Heating duration min	Average rate $^\circ\text{C}/\text{min}$		Index of volume growth	Specific energy consumption kWh/ kg
				Heating	Cooling		
1	585/ 567	873	55	15.5	5.8	2.20	1.13
2	585/ 565	870	56	15.2	5.6	2.20	1.16
3	585/ 568	862	58	14.5	5.9	2.30	1.19
4	585/ 569	855	57	14.7	5.7	2.30	1.17

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

Variant	Apparent density g/cm^3	Porosity %	Thermal conductivity $\text{W}/\text{m}\cdot\text{K}$	Compressive strength MPa	Water absorption %	Pore size mm
1	0.40	81.8	0.081	4.9	1.9	0.5 – 0.9
2	0.42	82.5	0.079	5.5	2.0	0.7 – 1.2
3	0.43	83.5	0.078	6.0	1.8	0.9 – 1.5
4	0.45	83.9	0.076	6.2	1.7	1.0 – 1.8

According to the contents of Table 2, the temperature range at which the foaming of the powder mixture occurred was between 855-873 °C, the highest value corresponding to the variant 1 with the lowest mass ratio of water glass (4%). So, the aqueous solution of water glass obviously influences the value of the foaming temperature of the material. The formation of silicates after the reaction between the water glass and the surface of the fine particles of glass waste as well as the release of the chemically bound water contained in the silicates facilitates the foaming process and this could be the main cause of the process temperature decreasing. Despite the reduction of the temperature, the duration of the process did not shorten, but remained relatively constant between 55-58 min. The specific energy consumption was also kept approximately constant at low values (1.13-1.19 kWh/ kg) compared to those achieved by conventional heating methods. Generally, the literature offers very few information about the specific energy consumption of the industrial processes. A market study [1] mentions that the Misapor consortium produces glass foam with an average energy consumption of 100 kWh/ m³ (i.e. up to 0.85 kWh/ kg), without counting the consumption for raw material processing. It should be considered that the Misapor's ovens use flat glass waste only in very low ratios, so that the energy consumption would be higher as the process temperature increases, probably exceeding 1 kWh/ kg. Considering the factors that influence the value of the specific consumption in an experimental microwave oven with discontinuous operation and of very small capacity as well as the energetic advantages offered by a microwave equipment of high capacity (it is considered a higher energy efficiency with up to 25 % [10], which would mean specific consumptions between 0.85-0.89 kWh/ kg), it turns out that, theoretically, the microwave field manufacturing process should be more efficient compared to a technological process based on conventional heating methods.

Analyzing the data in Table 3, it can be observed that the addition of water glass in the powder mixture of raw materials influences to a small extent the physical characteristics of the final product (apparent density, porosity and thermal conductivity) and more clearly the mechanical and morphological features. Thus, the value of the compressive strength increased from 4.9 up to 6.2 MPa and the pore size range in the section of the four samples also increased from 0.5-0.9 mm up to 1.0-1.8 mm with the water glass enrichment of the

mixture from 4 up to 13%. A closer examination of the variation of apparent density values of samples leads to the conclusion that the ceramic structure of the material becomes heavier as the increasing of water glass ratio in the mixture composition, although the pores size becomes larger. Thus, sample 4 having the largest volume occupied by pores has the highest apparent density. In contrast, the porosity values have a relatively normal variation, increasing with the increase of the pores volume. This variation would not be possible if the density of the ceramic structure was not also increasing.

Pictures of the longitudinal section of the cellular glass samples are shown in Figure 2. The images allow to observe a good homogeneity of the pore distribution in the section as well as the physical difference between the samples made with variable proportions of water glass.

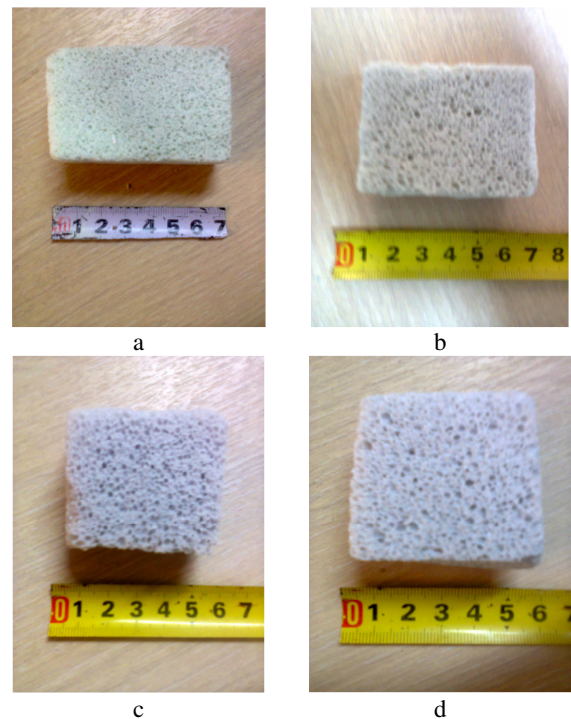
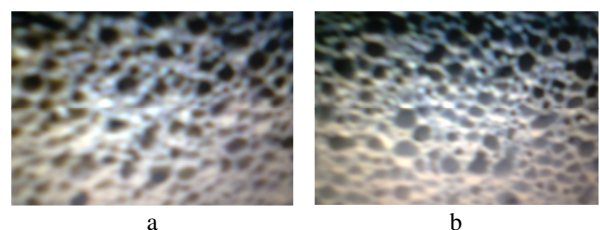


Figure 2. Pictures of the longitudinal section of the samples a – sample 1; b – sample 2; c – sample 3; d – sample 4.

The analysis of the microstructural configuration of the cellular glass samples was performed with a Smartphone digital microscope. The obtained images are shown in Figure 3.



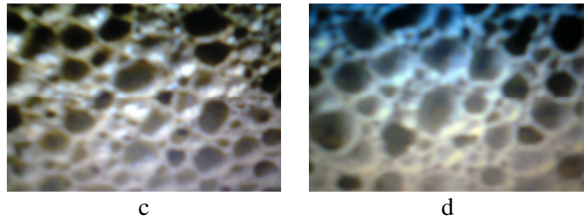


Figure 3. Images of the microstructural configuration of the samples
a – sample 1; b – sample 2; c – sample 3; d – sample 4.

The test for determining the hydrolytic stability was carried out according to the standard procedure ISO 719: 1985 (confirmed in 2011) with 0.15 ml of 0.01 M HCl solution. The results indicated that the stability of the foam glass gravel samples corresponds to the hydrolytic class no. 2.

3.2 Discussion

The main challenges in the manufacture of cellular glass from flat glass waste are obtaining a porous thermal insulating product, with an uniform distribution of the pores, so that the thermal protection provided by the material to be homogeneous as well as an increased energy efficiency of the manufacturing process. In the world, the flat glass waste is less interesting for the production of cellular glass, compared to other types of glass waste and especially, container glass waste. Previous experimental results showed that is possible a significant increase of the foaming process temperature and implicitly, of its duration, due to some contaminants existing in the raw material composition. The methods of eliminating these disadvantages, by adding some compounds (e.g. MnO_2) with the role of enhancing the oxidation of the powder mixture, generate an undesirable macrostructural inhomogeneity of the glass foam.

Experiments conducted in the last years in the company Daily Sourcing & Research Bucharest, using powder mixtures consisting of colorless flat glass waste, coal ash and silicon carbide as a foaming agent, placed inside a silicon carbide and silicon nitride enclosure with the wall thickness of 3.5 mm, introduced in a microwave oven, allowed the foaming temperature reduction from over 1000 °C up to 978 - 992 °C and the duration diminishing from 102 - 107 min up to 71 - 75 min, by intensifying the direct microwave heating. Also, the macrostructural homogeneity of the porous material was significantly improved. Own experiments involving the use of colorless flat glass waste and calcium carbonate as a foaming agent have not led to similar results regarding the macrostructural homogeneity of the foamed samples.

Therefore, the experimental results of this paper obtained in the microwave field manufacture of cellular glass with high mechanical strength and a good macrostructural homogeneity, represent an important and original contribution of the team of researchers from Daily Sourcing & Research.

4. CONCLUSION

The research, whose results are presented in the paper, aimed to produce by the microwave irradiation of a high mechanical strength cellular glass using colorless flat glass waste (between 86-95%), calcium carbonate (1%) as a foaming agent and a variable addition (between 4 - 13%) of sodium silicate called also "water glass".

The main challenges of the cellular glass production from flat glass waste are obtaining a thermal insulating product, with an uniform distribution of the pores and a higher energy efficiency of the microwave manufacturing process compared to the industrial conventional processes.

The oven used for the experiments was a 0.8 kW-domestic microwave oven adapted for high temperature operation (up to 1200 °C).

The ceramic tube made of SiC and Si_3N_4 in a 4/1 mass ratio, with the wall thickness of 3.5 mm, allowed both its penetration by the microwave and the contact with the powder mixture (direct microwave heating) and the absorption of the waves in the wall mass and the heat transfer by thermal radiation (indirect microwave heating). This mixed heating system proved to be optimal for rapidly reaching the required temperature in the material mass (between 855 - 873 °C) and did not affect its internal structural characteristic.

The characteristics of the cellular glass samples were the following: apparent density between 0.40 - 0.45 g/ cm^3 , porosity between 81.8 - 83.9%, thermal conductivity in the range 0.076 - 0.081 W/ m·K, compressive strength between 4.9 - 6.2 MPa (the maximum value corresponding to the maximum ratio of water glass) and water absorption below 2%, being suitable for their use as a foam glass gravel produced by conventional methods. The values of physical and mechanical properties are similar to those of the industrial products, although the flat glass waste used in experiments is, generally, avoided because it causes the structural inhomogeneity of the glass foam.

The pore size had restricted intervals for each investigated sample, their values being between 0.5 - 0.9 mm for sample 1 and between 1.0 - 1.8 mm for

sample 4. Thus, the objective regarding the homogeneity of the pore distribution been fulfilled.

The specific energy consumption had very low values (1.13 - 1.19 kWh/ kg) for a low power oven (0.8 kW) and with discontinuous operation. Generally, the information on this functional parameter of the conventional industrial ovens, is not published. Theoretically, according to the literature, a microwave equipment of high capacity, industrially usable, would allow an energy efficiency up to 25% higher compared to the oven experimentally used.

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