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Microwave Heat Treatment to Manufacture Foam Glass Gravel

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Abstract

The paper aimed at the experimental manufacture of a foam glass gravel type by sintering at over 900 ºC a powder mixture composed of recycled glass waste (92%), sodium borate (6%), kaolin (0.3%), silicon carbide (1.7%) and water addition (12%). The originality of the work was the application of the unconventional technique of microwave heating through a predominantly direct heating procedure. The product foamed at 908 ºC had a very fine porous structure (pore size between 0.05-0.20 mm) and a compressive strength above the usual level of foam glass gravels (7.8 MPa). The apparent density of 0.28 g/cm3 corresponding to a bulk density of 0.20 g/cm3 and the thermal conductivity of 0.075 W/m•K ensures the thermal insulating character of the material required for use in the specific field of applications of foam glass gravel. The manufacturing process had an excellent energy efficiency, the specific energy consumption decreasing up to 0.70 kWh/kg.

Introduction

New types of thermal insulation in the form of porous gravel with high compressive strength, with dimensions between 10-80 mm, are used as filler insulation material around the perimeter of buildings, under load-bearing foundations, insulating drainage layers of garden roofs, road and railway constructions, airport runway, bridge abutments, insulation of underground heating pipelines or storage tanks, etc. (Zegowitz, 2010; Cosmulescu et al., 2020).

The manufacturing technique of these foam glass gravels is simple involving the predominant use of recycled glass waste as raw material and some mineral additives in small proportions as foaming and fluxing agents (if appropriate). The finely ground and pressed mixture is heated to a high temperature in a conveyor belt oven, which facilitates the foaming process of the glass waste by releasing a gas into its softened mass with an adequate viscosity. The gas is captured as gas bubbles and by cooling, the bubbles turn into pores forming a porous structure (Scarinci et al., 2005).

Unlike the thermal insulation of buildings in the form of panels, for insulating at ground level, underground, underwater or as a roof garden of various constructions, there are special requirements regarding the compressive strength and resistance to water and frost of these materials. The advantages of a thermal insulation type of foam glass gravel are suitable for these applications (Zegowitz, 2010).

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In recent decades, with the beginning of the industrial-scale manufacture of glass foams from recycled glass waste, the production of foam glass gravels for the above mentioned applications has become a major concern of industrial manufacturers. The main European companies that produce foam glass gravel are Geocell Schaumglas (Austria), Misapor Switzerland (Switzerland), Glapor Werk Mitterteich (Germany), Veriso (Germany), Technopor Handels (Austria), Vetropor (Switzerland), Hasopor (Sweden), Glasopor (Norway), Foamit (Finland). Due to the difficult climatic conditions, the main market for foam glass gravel in the Scandinavian countries is represented by the road construction. The insulation of the asphalt layer from the frozen ground, the structural stability, the rapid drainage and the lack of capillary action prevent the degradation of the road caused by the freeze-thaw sequence (Cosmulescu et al., 2020).

One of the main European producers of foam glass gravel - Geocell Schaumglass (Geocell, 2018; Gafari et al., 2019) uses 90% colored post-consumer glass container and 10% colorless flat glass waste as raw material. The foaming agent is not specified. The sintering temperature is almost 900 ºC, the product being manufactured in tunnel oven with conveyor belt. Its main characteristics are: bulk density of 0.15 g/cm3, thermal conductivity of 0.080 W/m•K and compressive strength of 5.7 MPa.

Another important producer of foam glass gravel is Misapor Switzerland (Zegowitz, 2010; Technical, 2017; TECHNOpor, 2018). The manufacturing recipe includes 98% recycled glass waste (green container glass waste and other glass waste mixture) and 2% gypsum ($CaSO_4 \cdot 2H_2O$), limestone ($CaCO_3$) or silicon carbide (SiC) as foaming agents (separately used). The average sintering temperature of the raw material is 900 ºC. The final product has bulk density of 0.16-0.19 g/cm3 (compact material density of 0.21-0.25 g/cm3), thermal conductivity of 0.12 W/m•K and compressive strength of 4.9- 6.0 MPa. The water absorption can reach 6-10%.

Glapor Werk Miterteich, also a major manufacturer of foam glass gravel, uses in its manufacturing recipe a liquid foaming agent (glycerol) combined with a sodium silicate aqueous solution. Commonly, the weight ratios of the used materials are 87% recycled glass waste (flat glass or container glass waste), 1% glycerol, 12% sodium silicate. Additionally, less than 0.5% kaolin is added to the powder mixture. The bulk density of the product is between 0.13-0.21 g/cm3, the thermal conductivity is around 0.078 W/m•K and the compressive strength has values between 4.9-6.0 MPa (Glapor Schaumglasprodukte, 2017; Glapor, 2018). By using a liquid foaming agent, the foam glass gravel porosity is very fine with pore size below 300 μm.

It should be mentioned that the entire world production of foam glass gravel is made in industrial ovens that use conventional heating techniques (electrical resistances or burning fossil fuels). The energy consumptions of these manufacturing processes are not shown in the literature. The bibliographic sources (Hurley, 2003; Foam Glass, 2014) can be considered as exceptions, presenting average energy consumption values at the level of the entire company in glass foam manufacturing processes at Misapor and Energocell (Geocell Hungary) of 100 kWh/m3 and 140 kWh/m3, respectively.

A very energy efficient technique for heating solids is being tested in the last four years in glass foam manufacturing processes in the Romanian company Daily Sourcing & Research. The application of the unconventional microwave heating technique is based on the ability of some materials to absorb the microwave field and transform their power into heat by conversion. The thermal energy initiated in the core of material is very intense and it is transmitted through its entire volume to the peripheral areas (Jones et al., 2002). On the other hand, the microwave heating is selective by acting only on the material that is microwave

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susceptible (Kitchen et al., 2014). The other components of the microwave oven can remain almost cold if the thermal protection of the heated material is very good. Although known since the mid-20th century, the microwave heating has remained in an extremely low application stage. Until two decades ago, it was used only in drying and low temperature heating processes. Although it was experimentally found that several types of material (organics, ceramics, polymers, metals, glass, etc.) are suitable for an efficient microwave heating (Kharissova et al., 2010), the industrial application of this advanced procedure is still in a period of experimental testing.

One of the Romanian company concerns in recent years has also been the manufacture of foam glass gravel from glass waste using the microwave heating technique. The paper (Cosmulescu et al., 2020) makes a comparative analysis of four groups of technological variants of manufacture in microwave field of this type of product. Mixtures of container glass waste (green and amber) with calcium carbonate, sodium borate and sodium silicate (first group), glycerol, sodium silicate and water, the waste glass being a colorless flat glass (second group), same type of foaming agent and additives, but the glass waste being a mixture of colored container glass waste (third group) and as a last variant (Paunescu et al., 2020), a mixture of container glass waste of different colors (colorless, green and amber) in the 50/20/30 weight ratio and silicon carbide as a foaming agent were tested. The best experimental products were obtained by sintering at 855, 818, 823 and 922 ºC, respectively, having the density values of 0.62, 0.24, 0.24 and 0.35 g/cm3, respectively, the thermal conductivity values of 0.087, 0.063, 0.063 and 0.075 W/m•K, respectively and the compressive strength values of 7.4, 5.3, 5.9 and 7.5 MPa, respectively. The specific energy consumption of the microwave heating processes had quite low values of 1.07, 0.86, 0.88 and 1.00 kWh/kg, respectively.

Except for the characteristics of the samples in the first group, the other products were almost similar in terms of quality with industrially manufactured foam glass gravels. However, the characteristics and energy consumption of the product from the fourth group (using SiC as a foaming agent) could be improved and this desideratum was the objective of the current work.

Methods

The solution adopted by the authors was the use of silicon carbide (SiC) as a foaming agent. At temperatures above 900 ºC, SiC reacts with oxygen in the oxidizing atmosphere in the free spaces between the glass particles releasing carbon dioxide (CO_2), the gaseous compound that contributes to the foaming of powder glass and silica (SiO_2), which is incorporated into the glass, according to reaction (1).

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

Of all the possible chemical reactions of SiC (with oxygen, carbon monoxide or water vapor) reaction (1) has the most suitable thermodynamic conditions to occur at both 1000 K and 1500 K. The release of silica by the SiC oxidation may result in partial crystallization of the glass with cristobalite precipitation as a crystalline phase. Adding a very small proportion of kaolin to the glass powder is effective in reducing the cristobalite precipitation (Scarinci et al., 2005). Also, an addition of sodium borate (borax) ($Na_2B_4O_7 \cdot 10H_2O$) as a fluxing agent was adopted. Excepting the decrease of the sintering/foaming temperature, the sodium borate contributes to the increase of the mechanical, chemical and thermal shock resistance of the material due to the high boron content (11 wt.%) (Bernardo et al., 2007).

The experimental microwave equipment used by the Daily Sourcing & Research company is a 0.8 kW-microwave oven of the household type, but adapted for high temperature heating processes. The finely ground and previously pressed raw material is placed freely on a bed of

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ceramic fiber mattresses and insulated in the inner space of a ceramic tube from a microwave susceptible material (SiC + Si3N4 in a 80/20 weight ratio). The tube has an outer diameter of 125 mm, a height of 100 mm and a wall thickness of 2.5 mm. A ceramic lid of the same material closes the upper opening of the tube. A hole with a diameter of 30 mm provided centrally in the lid allows the visualization of the raw materials-additives mixture during the heating process. A very efficient thermal insulation from the ceramic fiber mattresses of the tube and lid inserted inside the microwave oven has the role of avoiding the heat loss outside the system, because the microwave heating is initiated in the core of the material mixture. The thickness of the 2.5 mm-ceramic tube was experimentally determined as optimal to reduce the intensity of the direct impact of microwaves with the glass-based raw material placed inside the tube. Partially, the tube wall absorbs the microwave field and heats up quickly transferring the heat inward through thermal radiation. This mixed microwave heating system has been adopted by the authors and used in all similar experiments in recent years. The temperature control of the heated material was performed with a radiation pyrometer mounted above the oven. The main components and the overall image of the microwave equipment are shown in Figure 1.

A B C

Figure 1. Experimental microwave equipment

A – 0.8 kW-microwave oven; B – ceramic fiber thermal protection;

C – SiC and Si_3N_4 ceramic tube.

The materials used in experiments were: post-consumer colorless, green and amber container glass (65/20/15 weight ratio), sodium borate and kaolin as raw material, silicon carbide as a foaming agent and water addition.

The recycled glass waste was broken, ground in a ball mill and sieved at a grain size below 100 μm. The chemical composition of the colorless, green and amber glass from the post-consumer container glass is shown in Table 1.

Table 1. Chemical composition of the glass waste

Chemical composition	**Glass waste type, wt.%**		
	Colorless	Green	Amber
SiO_2	71.7	71.8	71.1
Al_2O_3	1.9	1.9	2.0
CaO	12.0	11.8	12.1
Fe_2O_3	-	-	0.2
MgO	1.0	1.2	1.1
Na_2O	13.3	13.1	13.3
K_2O	-	0.1	0.1

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Cr_2O_3	0.05	0.09	-
TiO_2	-	-	0.05
SO_3	-	-	0.05

The sodium borate (borax) was used at a fluxing agent. Purchased from the market at a granulation below 400 µm, it was ground in an electrical laboratory device and sieved at a grain size below 100 µm.

The commercial kaolin, in the form of a very fine powder, was used as purchased on the market.

Also purchased from the market, the silicon carbide had a grain size below 6.3 µm.

In experiments, four experimental variants were adopted using different weight proportions of glass waste (between 91.9-92.0 wt.%), sodium silicate (between 5.7-6.0 wt.%) and silicon carbide (between 1.7-2.0 wt.%) as well as constant proportions of kaolin (0.3 wt.%) and water addition (12 wt.%). The composition of the experimental variants is shown in Table 2.

Table 2. Composition of the experimental variants

Variant	**Container glass waste wt.%**	**Silicon carbide wt.%**	**Sodium borate wt.%**	**Kaolin wt.%**	**Water addition wt.%**
1	92.0	2.0	5.7	0.3	12.0
2	91.9	2.0	5.8	0.3	12.0
3	91.9	1.8	6.0	0.3	12.0
4	92.0	1.7	6.0	0.3	12.0

Result and Discussion

The main functional parameters of the experimental manufacturing process of foam glass gravel presented in this paper are shown in Table 3.

Table 3. Main functional parameters of the manufacturing process

Parameter	**Variant 1**	**Variant 2**	**Variant 3**	**Variant 4**
Dry raw material/ foam glass gravel amount (g)	550/535	550/534	550/535	550/532
Sintering/foaming temperature (ºC)	920	916	912	908
Heating time (min)	49	42	39	36
Average rate (ºC/min) -heating -cooling	 18.4 6.6	 21.3 6.5	 22.9 6.7	 24.7 6.4
Index of volume growth (%)	1.80	1.70	1.55	1.40
Specific energy consumption (kWh/kg)	0.95	0.82	0.76	0.70

Due to the addition of the fluxing agent (sodium borate) in the powder mixture of the raw material, but also to the improvement of the thermal insulation conditions (with ceramic fiber mattresses resistant up to 1600 ºC) of the ceramic tube containing the heated material, the functional parameters of the heating process were optimized compared to the manufacturing process performed only with SiC as a foaming agent presented in the paper (Paunescu et al., 2020). Thus, the sintering/foaming temperature decreased from 916-929 ºC to 908-920 ºC, the process time was reduced from 43-55 min to 36-49 min, the heating rate reached the higher

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value of 24.7 ºC/min compared to only 20.9 ºC/min in the previous experiment and implicitly, the specific energy consumption was significantly reduced from 0.93-1.18 kWh/kg to 0.70-0.95 kWh/kg.

The appearance of the products corresponding to the four experimental variants is shown in Figure 2.

A

B

C

D

Figure 2. Appearance of the foam glass gravel samples

A – sample 1, sintered at 920 ºC; B – sample 2, sintered at 916 ºC;
C – sample 3, sintered at 912 ºC; D – sample 4, sintered at 908 ºC

The main characteristics of the experimentally obtained foamed products were determined by commonly used methods. The apparent density was measured by the gravimetric method (Manual, 1999) and the porosity was calculated by the method of comparing the true and apparent density (Anovitz & Cole, 2005). The guarded-comparative-longitudinal heat flow method (ASTM E1225-04 standard) was used to determine the thermal conductivity value. The water absorption of the foam glass gravel samples was measured by the water immersion method (ASTM D570 standard). A Digital Smartphone Microscope was used to observe the microstructural characteristics of the samples. To measure the compressive strength value of the foamed products a Stable Micro System TA XT Plus Texture Analyzer was used.

The physical, mechanical, thermal and morphological features of the foam glass gravel samples are presented in Table 4.

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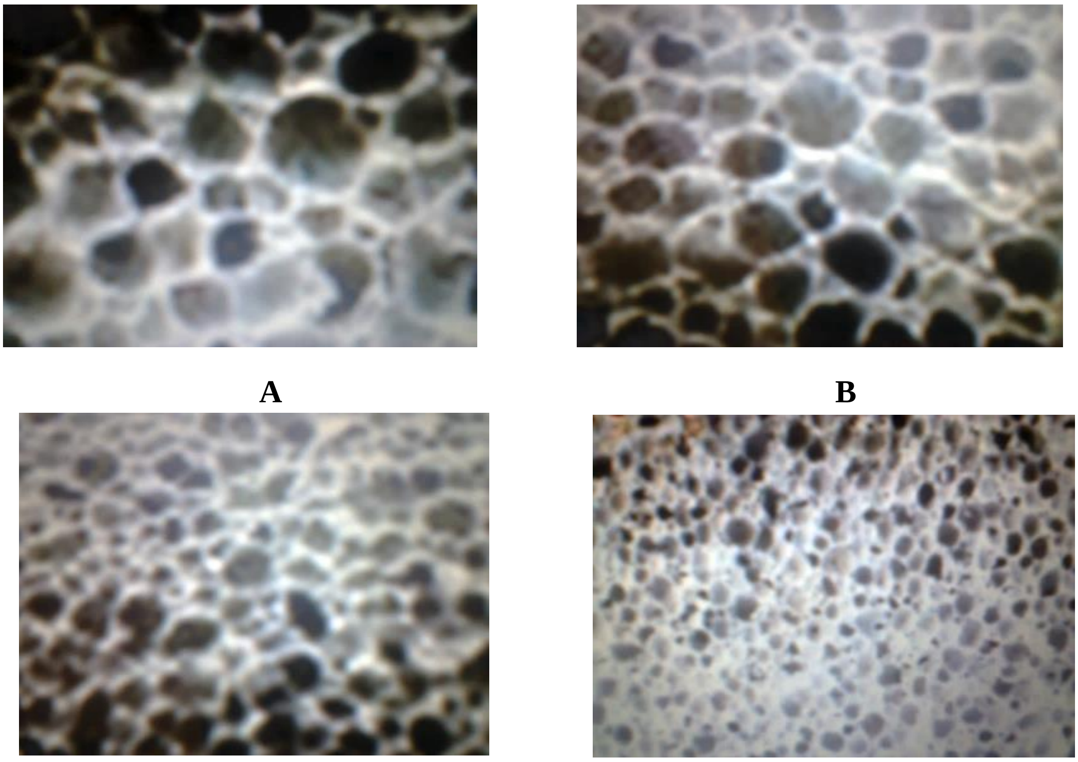
Table 4. Physical, mechanical, thermal and morphological characteristics of samples

Var.	Apparent density g/cm^3	Porosity %	Compressive strength MPa	Thermal conductivity W/m·K	Water absorption %	Pores dimension mm
1	0.38	82.7	6.0	0.089	2.1	0.35 – 0.70
2	0.34	84.5	6.8	0.084	1.6	0.25 – 0.55
3	0.31	85.9	7.4	0.080	1.5	0.10 – 0.40
4	0.28	87.3	7.8	0.075	1.3	0.05 – 0.20

According to the data in Table 4, the characteristics of the foam glass gravel samples are excellent for all four products obtained having the apparent density between 0.28-0.38 g/cm^3, porosity between 82.7-87.3%, thermal conductivity in the range of 0.075-0.089 W/m·K and compressive strength between 6.0-7.8 MPa, being appropriate in terms of quality for use as foam glass gravel in the fields of application mentioned above.

Examination of the data on product characteristics leads to the conclusion that the product corresponding to the experimental variant 4 is clearly the best. Obtained by sintering at only 908 ºC of the mixture consisting of 92.0 wt.% container glass waste, 6.0 wt.% sodium borate, 0.3 wt.% kaolin and 1.7 wt.% SiC, the optimal foam glass gravel sample has the apparent density of 0.28 g/cm^3, porosity of 87.3%, thermal conductivity of 0.075 W/m·K, compressive strength of 7.8 MPa, water absorption (for 1 hour immersion) of 1.3% and pore size between 0.05-0.20 mm.

The microstructural images of the four foam glass gravel products, including also the optimal product (sample 4) are shown in Figure 3. The pore distribution is homogeneous, the pore size decreasing with the decrease of the sintering/foaming temperature.



A **B**

C **D**

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Figure 3. Microstructural pictures of the foam glass gravel samples

A – sample 1; B – sample 2; C – sample 3; D – sample 4.

Usually, the characterization of industrially produced foam glass gravels is done by determining their bulk density. This involves measuring the mass of several pieces of foam glass gravel placed in a space with measured volume. In this way, the density calculation is influenced by the free spaces between the pieces which, in general, represent about 30% taking into account the average dimensions of the pieces (between 50-70 mm). Therefore, the bulk density of the experimentally manufactured samples whose apparent density determined individually taking into account the total pore volume was between 0.28-0.38 g/cm^3, could be in the range of 0.20-0.27 g/cm^3 almost similar to that of the industrially manufactured products, with the mention that the calculated bulk density is higher, instead the compressive strength reaches higher values.

By comparison, Geocell Schaumglas makes foam glass gravels with a bulk density of 0.15 g/cm^3, thermal conductivity of 0.08 W/m·K and compressive strength of 5.7 MPa (Geocell, 2018; Gafari et al., 2019). Misapor produces the same type of products with the bulk density between 0.16-0.19 g/cm^3, thermal conductivity of 0.12 W/m·K and compressive strength between 4.9-6.0 MPa (Technical, 2017; TECHNOpor, 2018). Glapor Werk Mitterteich, which uses a liquid foaming agent (glycerol), obtains products with the bulk density of 0.13-0.21 g/cm^3, thermal conductivity of 0.078 W/m·K and compressive strength between 4.9-6.0 MPa (Glapor Schaumglasprodukte, 2017; Glapor, 2018).

In energy terms, the comparison of the optimal product of foam glass gravel experimentally obtained with similar products industrially manufactured cannot be made due to the lack of information in the literature. The specific energy consumption (0.70 kWh/kg) corresponding to the optimal sample (variant 4) reported to the calculated bulk density of 0.20 g/cm^3 (or 200 kg/m^3) leads to the value of 140 kWh/m^3. This value coincides with the average specific consumption of Geocell Hungary company, but this does not provide real indications of the energy consumption.

The only reports in the literature of the specific energy consumption belong to the company Daily Sourcing & Research. All experiments in the field of foam glass gravel manufacturing were performed with the unconventional microwave heating technique and the values of specific energy consumption were between 0.86-1.07 kWh/kg (Paunescu et al., 2020). Compared to these previously obtained values, the specific consumption of the sample made with the experimental variant 4 (0.70 kWh/kg) is clearly the most economical.

Conclusion

In recent decades, the production of foam glass gravel has become a major concern of industrial manufacturers. Geocell, Misapor and Glapor are the main European companies that produce this glass foam type for insulations in special conditions by conventional techniques of heat treatment. The Romanian company Daily Sourcing & Research has experimentally made in recent years different types of foam glass gravel with physical, thermal, mechanical and morphological characteristics almost similar to those industrially manufactured by the unconventional technique of microwave heating, which is the originality of the work. The last achievement presented in the paper was a porous product with very high compressive strength (up to 7.8 MPa) compared to other existing foam glass gravel using a mixture composed of recycled glass waste (92%), sodium borate (6%), kaolin (0.3%), silicon carbide (1.7%) and water addition (12%). By sintering at 908 ºC, a product with the apparent density of 0.28 g/cm3 (bulk density of 0.20 g/cm3), porosity of 87.3%, thermal conductivity of 0.075 W/m•K, water absorption of 1.3% and pore size between 0.05-0.20 mm was obtained. The specific energy consumption was very low (0.70 kWh/kg).

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