COMPARATIVE ANALYSIS OF THE FOAM GLASS GRAVEL TYPES EXPERIMENTALLY PRODUCED BY MICROWAVE IRRADIATION

FELICIA COSMULESCU[1], LUCIAN PAUNESCU[*2], MARIUS FLORIN DRAGOESCU[2], SORIN MIRCEA AXINTE[2, 3]

[1]*Cosfel Actual SRL, 95-97 Calea Grivitei, sector 1, Bucharest 010705, Romania*

[2]*Daily Sourcing & Research SRL, 95-97 Calea Grivitei, sector 1, Bucharest 010705, Romania*

[3] *University "Politehnica" of Bucharest, Faculty of Applied Chemistry and Materials Science, 1-7 Gh. Polizu street, sector 1, Bucharest 011061, Romania*

Abstract: The paper presents an unconventional technique for manufacturing foam glass gravel. The numerous experimental results obtained by the authors in the last year using various manufacturing recipes common in the world industrial production by conventional techniques were selected and subjected to a qualitative and energy efficiency comparative analysis, aiming to determine the best foam glass gravel manufacturing procedure under the conditions of the use of the microwave energy. The optimum recipe was composed of 83% glass waste, 1 % glycerol, 8 % sodium silicate and 8 % water. The maximum heating temperature reached 823 ºC with a very economical energy consumption of 0.88 kWh/kg. The sample characteristics were apparent density-0.24 g/cm^3, porosity-89.1% with pore size between 0.3 - 0.6 mm, thermal conductivity-0.063 W/m·K and compressive strength - 5.9 MPa, almost similar to those industrially obtained.

Keywords: foam glass gravel, glass waste, microwave, compressive strength, specific energy consumption, comparative analysis

1. INTRODUCTION

In the last decades of the 20th century, the waste recycling of plastic, metal, glass, paper, etc. with high annual generation rates has become a common practice in the world, especially in developed and developing countries. Glass recycling also follows the same trend. In the case of glass waste, recycling is focused primarily on the use of waste as a raw material for the industrial manufacture of the new glass. However, the high costs of processing and separation of the color glass waste for reuse in the glass industry have provided the considerably cheaper variant of the manufacture of cellular glass from glass waste usable in building as a substitute for some traditional building materials on the market [1]. The manufacture of the cellular glass by thermal treatments at high temperature began in the second half of the last century. Initially, it was not used glass waste, but pristine glass [1]. Different sorts of cellular glass are available for various requirements, from lightweight porous materials used especially as insulating materials, up to porous materials with high mechanical strength (generally, up to 6 MPa) used as filling material in constructions that require mechanical stress [1-3]. The foam glass gravels represent such products with porous structure, light weight, low thermal conductivity, and

* Corresponding author, email: lucianpaunescu16@gmail.com

simultaneous, high compressive strength. The manufacture of foam glass gravel uses container glass and flat glass waste. The cullet is crushed and processed to remove the gross contamination of metal, labels, plastics, cork, etc. Then, the processed cullet is dried to remove water and reduce the organic carbon contain to a minimum allowed limit (0.5%). The dried cullet is ground in a ball mill and sieved below 100 μm. The foaming agent is added in the glass powder and mixed. Each company has the own manufacturing recipe [3]. Several types of foam glass gravel using different manufacturing recipes are currently produced in some European countries. The main industrial manufacturers of foam glass gravel are Geocell Schaumglas (Austria), Misapor Switzerland (Switzerland), Glapor Werk Mitterteich (Germany), Veriso (Germany), Technopor Handels (Austria), Hasopor (Sweden), Glasopor (Norway), Foamit (Finland), Vetropor (Switzerland). In the Scandinavian countries the main market for foam glass gravel is represented by the road construction due to the difficult climatic conditions. 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.

Geocell Schaumglas is one of the leading European manufacturers of foam glass gravel, having four facilities in Austria and Germany [4]. The raw material is constituted by recycled glass (90 wt.% colored post-consumer container glass and 10 wt.% colorless flat glass waste). The mineral activator used in mixing with the raw material is not specified. The sintering-foaming process is carried out in tunnel furnaces with conveyor belt at temperatures of almost 900 ºC. Foam glass gravel has a porous structure with closed cells, the bulk density of 0.15 g/cm^3, the thermal conductivity of 0.08 W/m·K and the compressive strength of 5.7 MPa. The product is extremely versatile due to its unique features of light weight, load bearing, draining and insulating properties. Also, it is fireproof and resistant to various external agents. The Geocell foam glass gravel is a suitable replacement for traditional building materials in: insulation of basements-under the slab and rooftops, new floor in old buildings, lightweight filling material for landscaping, roof gardens, green roofs, road and railway constructions, bridge abutments, insulation of underground pipelines.

Misapor Switzerland produces foam glass gravel in two facilities in Switzerland, one in Italy and one in Germany. Since 2009, Misapor has operated as an international group of companies [5]. For manufacturing foam glass gravel, Misapor used 98 wt.% recycled glass composed of mixed glass waste and green container glass waste as well as 2 wt.% gypsum, limestone or silicon carbide as foaming agents individually used. The heating process carried out in conveyor furnaces at an average temperature of 900 ºC. The duration of the material passage over the entire length of the oven is about 30 minutes. The Misapor foam glass gravel has insulating, drainage, load bearing characteristics, is very light, frost-proof, freeze-thaw resistant [6]. The product has the bulk density of 0.16-0.19 g/cm^3 and the compact material density of 0.21-0.25 g/cm^3. The thermal conductivity of the compact material is 0.12 W/m·K and the coefficient of water absorption can reach 6-10%. The compressive strength has high values between 4.9-6.0 MPa. Currently, it is used in building and civil engineering for insulation, as lightweight aggregate for drainage, noise barriers, in the garden and landscaping, etc. Recently, Misapor was focused only for the perimeter insulation products [7].

The German company Glapor Werk Mitterteich is also specialized to manufacture foam glass gravel [8, 9]. The manufacturing recipe includes 87 wt.% recycled glass (flat glass or container glass waste), 1 wt.% glycerol, 12 wt.% sodium silicate and below 0.5 wt.% kaolin. The bulk density of this product has values between 0.13-0.21 g/cm^3 and the compressive strength is between 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.

Although the German company Glamaco is only a very important supplier of industrial equipment for the manufacture of foam glass gravel, the data from literature on manufacturing recipes, product characteristics and the fields of application are much richer compared to those of others companies. The furnace type provided by Glamaco is a tunnel furnace with conveyor belt powered by natural gas burning. The heating process can be direct or indirect. The maximum foaming temperature is 900 ºC. Scrap or glass waste are used as raw material and silicon carbide, manganese oxide, glycerol, sodium silicate, coal powder (or soot), calcium carbonate, gypsum are the mineral activator materials used as foaming agents. One of the manufacture recipe provided by the literature contains 95 wt.% glass waste (with the grain size below 100 μm), glycerol and calcium carbonate as foaming agents as well as sodium silicate as an enveloping material, accounting for 5 wt.%, water addition and very low kaolin ratio. To obtain 1 m^3 glass foam it is necessary about 150-200 kg glass-based raw material. The main features of the foam glass gravel produced in furnaces provided by Glamaco are: density of the compact material between 0.15-0.20 g/cm^3, bulk density between 0.10-0.15 g/cm^3, thermal conductivity in the range 0.06-0.08 W/m·K and compressive strength between 4-6 MPa. The products are used as an insulating material for several applications: insulation of platform and terrace roofs, green roof thermal insulating with drainage function, insulating for special applications (swimming pools, tunnels), substructure in road and railway constructions, insulating for long-distance heating pipelines and underground storage tanks [10, 11].

Refaglass Trade is a Czech producer of lightweight drainage gravel, insulation of foundation slabs, floors and roofs [12]. Its products are made of recycled glass waste. The literature does not provide information about manufacturing recipes and features of the foam glass gravel.

The Swedish company Hasopor produces foam glass gravels with dimensions between 10-60 mm from recycled container glass. The powder raw material is mixed with an unspecified mineral activator. The heating process is carried out in a conventional tunnel furnace at about 1000 ºC, the foamed material growing in volume 4-5 times. The bulk density of the foam glass gravel is about 0.18 g/cm^3. The products of Hasopor company are mainly used as a lightweight filling material for road construction. Also, they are used for house and geotechnical construction areas [13].

The production of the Norwegian company Glasopor is specialized in the manufacture of foam glass gravels intended mainly for drainage and insulation in the field of road construction. The porosity of the material is around 80% and the bulk density is 0.18 g/cm^3 [14].

The foam glass gravel manufactured by the Finnish company Foamit is a lightweight and insulating material, which can be used in a large range of structural solutions for infrastructure construction and house building. It is made of clean recycled glass. The foam glass is non-flammable and has good water conductivity properties. Due to its cellular structure the foam glass gravel allows an excellent thermal insulation. The piece's dimension varies from 3-10 mm up to a maximum of 60 mm. The bulk density of this glass foam is between 0.18-0.21 g/cm^3 and the thermal conductivity has values in the range 0.08-0.13 W/m·K [15].

According to the literature, all industrial production of foam glass gravel worldwide is based exclusively on conventional heating techniques (burning fossil fuels or electrical resistances). An advanced heating method based on the unconventional technique of microwave using has been known since the mid-20th century [1], but its use has been limited to only a few applications, of which household food preparation is well known. At the industrial level, the use of microwaves is still extremely limited, although in the last 10-15 years the research has found that the area of applicability could be extended to many kinds of materials (organics, ceramics, metals, glass, polymers, etc.) [16].

A paper published in 1997 [17], showed that due to its rich composition in SiO_2 and Al_2O_3, compounds that are not microwave susceptible, the glass cannot be efficiently heated in the microwave field starting at the ambient temperature, but only above 500 ºC, when the dielectric characteristics of this material strongly improve allowing the absorption of the electromagnetic energy and its conversion into heat. This conclusion, theoretically correct, was taken over by other authors in their works [1, 2], decisively influencing the attitude of researchers and manufacturers in the glass industry towards the possibility of producing glass foam exclusively by microwave irradiation. Practically, however, the commercial glass representing the base of the glass waste used in this process inherently contains some contaminants (Fe_2O_3, Cr_2O_3, etc.), which are microwave susceptible and allow a rapid and efficient heating even from the ambient temperature, fact experimentally proven by tests performed in the Romanian company Daily Sourcing & Research Bucharest [18]. Thus, all the research carried out in this company in the field of glass foam production has been focused on the use of microwaves.

Several experimental results were obtained by the authors of the current paper in the last year using various manufacturing recipes of foam glass gravel through the own microwave heating technique. These recipes were included in four distinct groups by the nature and proportions of the foaming agent and additives as well as the process parameters: (a) based on calcium carbonate, borax and sodium silicate; (b) based on glycerol, sodium silicate and water, the glass waste being colorless flat glass; (c) based on the same type of foaming agent and additives, but the glass waste being a mixture of container glass waste; (d) based on silicon carbide. From each group the optimal variant of foam glass gravel was selected, and the paper constituted a comparative analysis of their qualitative performance and energy consumption.

2. METHODS AND MATERIALS

2.1. Methods

The basic principle of producing a cellular glass from glass waste is the heating of the raw material powder including also a foaming agent until the softening temperature of the glass is reached. The foaming agent, either solid or liquid, must be so adopted that there is a correlation between the temperature at which the decomposition or oxidation reaction of the agent occurs with the release of a gaseous compound and the softening temperature of the raw material, so that the gas can meet a sufficiently viscous material and to be blocked like bubbles in its mass. By cooling, the bubbles will form a homogeneous porous structure. There are numerous factors that influence the quality of the cellular glass: the temperature and rate of heating, the maintaining time at the foaming temperature, the weight ratio of the foaming agent, the nature and the ratio of other mineral additions with the role to improve the process parameters, etc. [1]. The foam glass gravel is a cellular glass type with a homogeneous cellular structure to ensure a good thermal insulation and with low pore size, so that the compressive strength of the material to be high enough, considering that the applicability area of foam glass gravel requires resistance to mechanical stress. Carbon-containing liquid foaming agents (e.g. glycerol) are indicated for the manufacture of foam glass gravel because the liquid penetrates much more easily into the free spaces between the fine glass particles as compared to a solid agent with a very fine granulation. The glycerol ($C_3H_8O_3$) decomposes in the oxidizing atmosphere of the furnace forming a wide range of compounds between pure carbon and CO_2 as well as hydroxyl compounds [19]. The carbon, in turn, oxidizes by releasing CO_2 and/ or CO. Due to the internal pressure, the material increases its volume by expansion. In order, to avoid the premature burning of carbon, in this case an aqueous solution of sodium silicate is used with the role of enveloping the glass particles.

A very effective and commonly used solid foaming agent is the silicon carbide (SiC). It oxidizes to over 900 ºC [20] resulting CO_2 and CO, which contribute to the glass foaming as well as SiO_2 and SiO, which enter in the glass composition, according to reactions (1) and (2).

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

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

The calcium carbonate ($CaCO_3$) is also a very commonly used foaming agent. The reaction by which a gas (CO_2) is released contributing to the foaming process is the thermal decomposition at temperatures above 800 ºC of the carbonate [1], according to the reaction (3). The calcium oxide enters in the glass composition.

$$CaCO_3 = CaO + CO_2 \quad (3)$$

In order, to obtain a dense foamed material with high mechanical strength, borax can be used as a fluxing agent and sodium silicate aqueous solution as a binder. The introduction of sodium silicate into the glass waste contributes to the homogenization of its chemical composition [21].

These manufacturing recipe types were experimented on the microwave oven and presented comparatively in this paper.

Using the microwave field in the process of sintering-foaming the glass waste, an important problem was canceling the destructive effect on the internal structure of the glass caused by the extremely intense exclusively direct contact of the waves with the material [18] through a branched distribution of the electromagnetic waves radiation. The external protection of the material subjected to heating with a ceramic screen (crucible or tube with a lid) made of a silicon carbide-based high microwave susceptible material has experimentally proven to be the optimal solution adopted by the team of authors. The wall thickness of the screen is essential (between 3.5-5 mm) to ensure an adequate mixed heating, partially direct and partially indirect, by absorbing a part of the microwave radiation in the screen mass which heats up quickly and intensely and transfers thermal energy through radiation. The experimental microwave equipment was a 0.8 kW-oven currently used in the household equipped with an only microwave generator adapted for high temperature operation (over 1200 ºC). The previously pressed material was placed on an insulating bed made of ceramic fiber and the ceramic crucible/tube is protected on the outside with ceramic fiber mattresses to prevent the heat loss. The temperature control of the material during the process was performed with a radiation pyrometer mounted above the oven, which can visualize the surface of the material through holes provided on the viewing axis. The experimental microwave equipment is shown in Figure1.

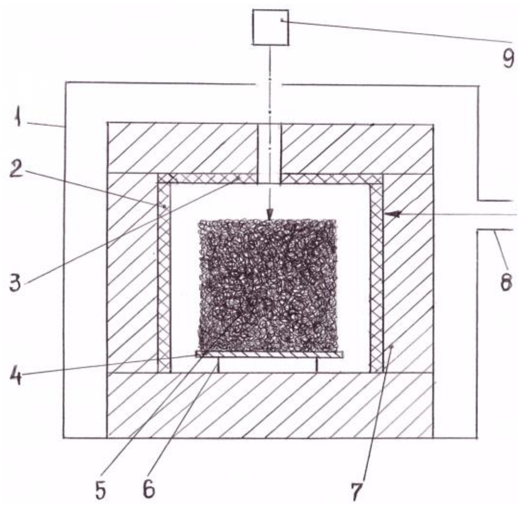


Fig. 1. Constructive scheme of the experimental microwave equipment:1 – 0.8 kW-microwave oven; 2 – ceramic tube; 3 – ceramic lid; 4 – metal plate; 5 – pressed powder mixture; 6 – metal support; 7 – ceramic fiber mattress; 8 – microwave source; 9 – radiation pyrometer.

2.2. Materials

The materials used in the experiments presented in the paper are: container glass (colorless, green and amber) waste, flat glass waste, calcium carbonate, glycerol, silicon carbide, borax and sodium silicate. The chemical composition of the glass waste kinds is shown in Table 1.

Table 1. Chemical composition of different kinds of glass waste.

Chemical composition	Container glass (wt.%)			Flat glass (wt.%)
	Colorless	Green	Amber	
SiO_2	71.7	71.8	71.1	71.1
Al_2O_3	1.9	1.9	2.0	1.3
CaO	12.0	11.8	12.1	9.3
Fe_2O_3	0.05	0.01	0.25	0.2
MgO	1.0	1.2	1.1	3.9
Na_2O	13.3	13.1	13.3	Total 14.2
K_2O	-	0.1	0.1	
Cr_2O_3	0.05	0.09	-	-
SO_3	-	-	0.05	-

The glass waste was thermally washed in a own conception microwave oven at 250 ºC to remove the inherent organic contaminants. The clean glass was ground in a ball mill and sieved at a grain size below 250 µm.

The calcium carbonate ($CaCO_3$) and silicon carbide (SiC) were used as solid foaming agents at grain size values below 40 µm without other mechanical processing such as they were purchased from the market.

The glycerol as a liquid carbonic foaming agent was used in association with a 30% aqueous solution of sodium silicate and water for the complete dissolution of glycerol and sodium silicate. The preparation of the liquid mixture was carried out separately before mixing it with the glass waste powder.

Due to the high Na_2O content, the borax was used as a fluxing material in the mixture with the glass powder. Also, by its important boron content (about 11 wt.%), it contributes to increasing the mechanical strength of the final product whose component it is.

The sodium silicate (Na_2SiO_3), also called "water glass", was used as a binder and adhesive. Its contribution to increasing the mechanical strength is remarkable [22].

2.3. Characterization of the foam glass gravel samples

The foam glass gravel samples experimentally obtained by the sintering/ foaming process of glass waste were characterized by traditional analysis methods. The main physical, mechanical and morphological characteristics were: apparent density, porosity, thermal conductivity, compressive strength, water absorption and microstructural configuration of the samples. The apparent density was measured by the gravimetric method [23]. The porosity was calculated by the comparison method [24] between the porous sample density and the density of the same material type in compact state obtained by melting followed by cooling to the room temperature. The determination method of the thermal conductivity [25] consisted of measuring the thermal flow value that passes through a sample of standard dimensions (50 mm-thickness) placed between two metal plates. One of the plate was heated and protected with insulating material and the other was cooled. An own conception device was used to determine the compressive strength by developing an axial pressing force generated with a hydraulically operated piston. The last pressing force axially applied to the sample before to crack was considered the compressive strength value. The tested sample had a cylindrical shape with the diameter of 80 mm and the height of 70 mm. The water absorption of the porous sample was measured by the traditional method of its water immersion (ASTM D 570). The porous microstructure of the foam glass gravel samples was identified with a Smartphone digital microscope.

Generally, for the physical characterization of foam glass gravel the literature used the bulk density, characteristic of some samples with irregular shapes and relatively small dimensions (up to 60-80 mm). For this determination, obviously, it would have required a great number of pieces occupying a closed space with known volume, which should have been weighed. The ratio between mass and volume, which includes many free spaces between pieces, represents the bulk density. In the case of the experiments described in the paper, although several tests were carried out under similar conditions to ensure the veracity of the results, there were not enough samples that could be used to determine a bulk density. In this situation, the apparent density was measured and shown in the tables, although its value is clearly greater than the bulk density.

3. RESULTS AND DISCUSSION

3.1. Results

Numerous experiments have been carried out in the last year applying several manufacturing recipes of foam glass gravel, the thermal treatment being achieved by the same unconventional technique of using the microwave radiation. Four groups of tests were performed clearly differentiated by the type of foaming agent, the nature of the mineral additions and the kind of glass waste used.

The first group of tests included the use of calcium carbonate ($CaCO_3$) as a foaming agent (1.5 wt.%), borax as a fluxing agent (between 3.0-8.0 wt.%) and an aqueous solution of sodium silicate (3.0-8.0 wt.%) as a binder and adhesive. A mixture of colored container glass (green and amber) in weight proportion between 83.5-89.5 wt.% was used as raw material. The maximum process temperature varied between 835-855 ºC, its duration being between 38-44 min. The specific energy consumption had values in the range 0.92-1.07 kWh/kg. The characterization of the foam glass gravel samples led to the following results: apparent density between 0.45-0.80 g/cm^3, porosity between 63.6-79.5%, thermal conductivity in the range 0.071-0.105 W/m·K, compressive strength between 3.5-9.5 MPa and the pore size from 0.8-1.0 mm up to 2.0-3.5 mm.

The second experiments group was based on the use of glycerol (1.0-1.8 wt.%) associated with sodium silicate (between 5.3-7.5 wt.%) and water (between 7.7-10.0 wt.%). Colorless flat glass waste (between 83.0-83.7 wt.%) was used as raw material. The functional parameters were: sintering/ foaming temperature (810-824 ºC), process duration (39-42 min) and the specific energy consumption (0.81-0.88 kWh/kg). The manufactured samples had apparent density between 0.20-0.26 g/cm^3, porosity between 85.5-88.2%, thermal conductivity in the range 0.056-0.070 W/m·K, compressive strength between 4.6-5.8 MPa and pore size from 0.3-0.8 mm up to 0.8-1.1 mm.

The third tests group was almost similar to the previous, the major difference being the use of a mixture of container glass waste (colorless, green and amber in the 50/20/30 ratio) between 82.3-83.0 wt.%. The glycerol varied between 1.0-1.6 wt.%, the sodium silicate was between 8.0-10.1 wt.% and the water between 6.0-8.0 wt.%. The process parameters were: the temperature between 815-823 ºC, the duration between 40-42 min and the specific energy consumption between 0.83-0.88 kWh/kg. The foam glass gravel had apparent density between 0.21-0.24 g/cm^3, porosity between 89.1-90.5%, thermal conductivity in the range 0.057-0.063 W/m·K, compressive strength between 4.8-5.9 MPa and pore size from 0.3-0.6 mm up to 0.7-0.9 mm.

The fourth group of experiments was based on a constant manufacturing recipe consisting of 84.9 wt.% container glass waste (colorless, green and amber), 1.7 wt.% silicon carbide (SiC) as a foaming agent and 13.4 wt.% water addition as a binder. The functional parameters of the process were: foaming temperature between 916-929 ºC, duration between 43-55 min and specific energy consumption between 0.93-1.18 kWh/kg. The products had apparent density between 0.31-0.40 g/cm^3, porosity between 77.7-83.6%, thermal conductivity in the range 0.063-0.089 W/m·K, compressive strength between 5.3-8.0 MPa and pore size from 0.4-0.7 mm up to 0.9-1.9 mm.

From each group of experiments the most representative variant of producing foam glass gravel was selected. Thus, a group consisting of the best four selected samples has been set up, which will be compared further. Table 2 contains the composition of the four selected variants and Table 3 and 4 shows the functional parameters of the manufacturing process and the main physical, mechanical and morphological features of the selected samples, respectively.

Table 2. Composition of the experimental variants (wt.%).

Var.	Container glass waste			Colorless flat glass	$CaCO_3$	Borax	SiC	Na_2SiO_3	Glycerol	Water
	Colorless	Green	Amber							
1	-	87.5		-	1.5	3.0	-	8.0	-	-
2	-	-	-	83.7	-	-	-	5.9	1.3	9.1
3	83.0			-	-	-	-	8.0	1.0	8.0
4	84.9			-	-	-	1.7	-	-	13.4

Table 3. Main functional parameters of the sintering-foaming process.

Variant	Wet 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	570/ 548	855	44	19.0	5.4	1.50	1.07
2	665/ 557	818	41	19.5	5.7	1.70	0.86
3	672/ 558	823	42	19.1	6.1	1.65	0.88
4	646/ 545	922	47	19.2	6.0	1.83	1.00

Table 4. Main physical, mechanical and morphological features of samples

Variant	Apparent density (g/cm^3)	Porosity (%)	Thermal conductivity (W/m·K)	Compressive strength (MPa)	Water absorption (%)	Pore size (mm)
1	0.62	63.6	0.087	7.4	7.4	1.0 – 1.6
2	0.24	86.4	0.063	5.3	1.4	0.5 – 0.9
3	0.24	89.1	0.063	5.9	0.6	0.3 – 0.6
4	0.35	80.9	0.075	7.5	0.4	0.5 – 1.0

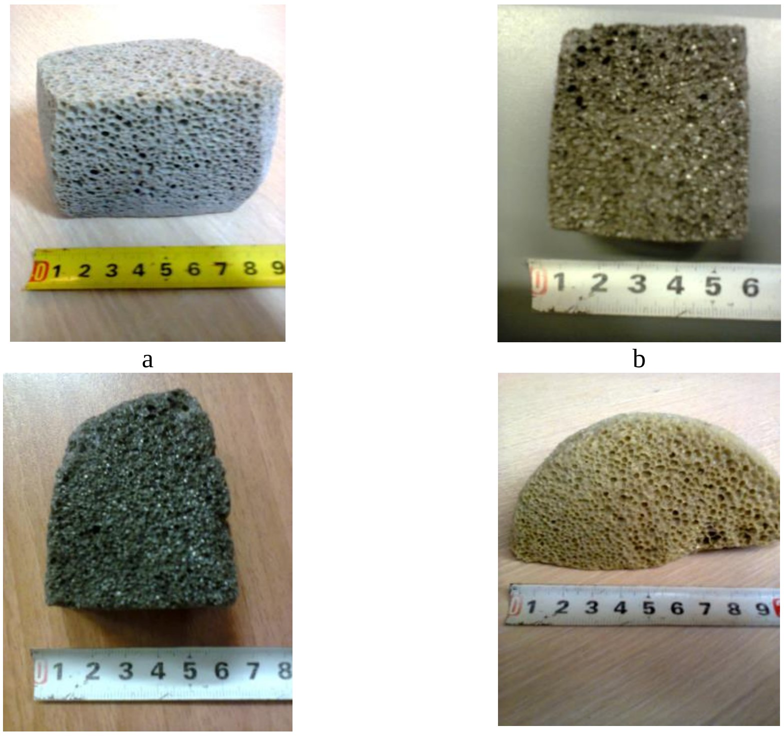
Therefore, according to the Table 2, two manufacturing variants using glycerol as a liquid foaming agent (variants 2 and 3) and two variants using solid foaming agents (calcium carbonate - variant 1 and silicon carbide - variant 4) should be compared. The main difference between variants 2 and 3 consists of the type of glass waste used: flat glass waste - variant 2 and container glass waste (colorless, green and amber) - variant 3.

From the data presented in Table 3, it results that the use of glycerol as a liquid foaming agent (variants 2 and 3) required the lowest temperatures (818-823 ºC) and durations (41-42 min) of the process of foaming the glass waste leading implicitly, to the most economical energy consumptions (0.86-0.88 kWh/kg). The manufacturing process of foam glass gravel with silicon carbide as a foaming agent and without other fluxing agents (variant 4) required the highest value of the foaming temperature (922 ºC). Also, the process duration was the longest (47 min) and required a specific energy consumption of 1 kWh/kg. By comparison, a manufacturing recipe of a dense cellular glass (variant 1), which is not explicitly mentioned in the literature in the industrial manufacturing processes of foam glass gravel, required a heating temperature of 855 ºC reached in 44 minutes, the specific energy consumption being 1.07 kWh/kg.

According to the data in Table 4, the lowest values of the apparent density (0.24 g/cm^3) and the thermal conductivity (0.063 W/m·K) of the foam glass gravel samples correspond to the variant 2 and 3. The porosities of the samples made by the two variants had high values (86.4 and 89.1%) and were characterized by homogeneous structures with closed pores with dimensions between 0.3-0.6 mm (variant 3) and 0.5-0.9 mm (variant 2). Sample 4 obtained with silicon carbide had slightly higher values of the apparent density (0.35 g/cm^3) and the thermal conductivity (0.075 W/m·K), the porous structure of material being very homogeneous with pore size between 0.5-1.0 mm. Sample 1 foamed with calcium carbonate as a foaming agent, borax and sodium silicate as additives, clearly differs from the other samples by the much higher apparent density (0.62 g/cm^3) and the much lower porosity (63.6%) compared to the usual physical characteristics of industrially manufactured foam glass gravels. The material had a homogeneous structure with low pore size between 1.0-1.6 mm.

The compressive strength of the two samples foamed with glycerol had the lowest values from the batch of experimentally produced samples, being 5.3 MPa (sample 2) and 5.9 MPa (sample 3), respectively, but these values are in the limits indicated in the literature (4-6 MPa) for the best foam glass gravels industrially manufactured. Sample 4 had the highest value of the compressive strength (7.5 MPa) with about 25% above the maximum limit required in the common applications of this product type, but the high level of the mechanical characteristic has affected its physical features. The compressive strength of sample 1 had a very high value (7.4 MPa), similar to sample 4, but the fact that the porous product had an apparent density well above the values recommended in the literature eliminated the possibility of its use as a quality foam glass gravel.

Pictures of the four samples selected for the comparative analysis are shown in Figure 2 and pictures of their microstructural configuration are shown in Figure 3. As an aspect, all the samples shown in Figure 2 are similar to industrially manufactured foam glass gravel pieces.



a b

c d

Fig. 2. Pictures of the experimental samples: a – sample 1, heated at 855 ºC; b – sample 2, heated at 818 ºC; c – sample 3, heated at 823 ºC; d – sample 4, heated at 922 ºC.

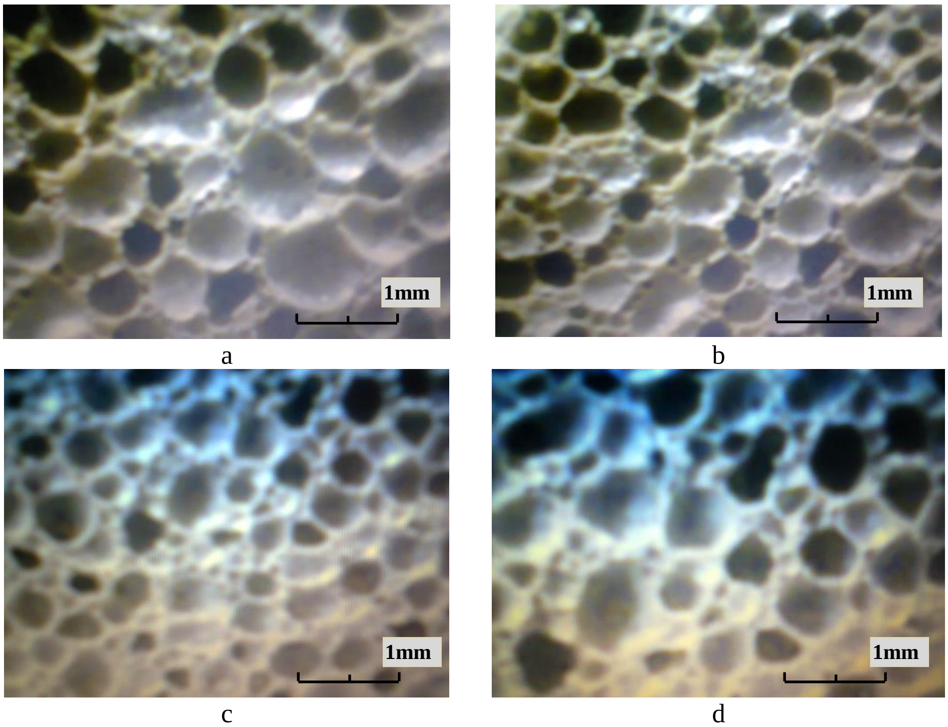


Fig. 3. Pictures of the porous microstructure of the samples: a – sample 1, heated at 855 ºC; b – sample 2, heated at 818 ºC; c – sample 3, heated at 823 ºC; d – sample 4, heated at 922 ºC.

3.2. Discussion

Therefore, the foam glass gravel samples experimentally obtained by microwave irradiation using glycerol as a foaming agent in association with 30% aqueous solution of sodium silicate and water correspond in the greatest extent to the requirements for the use in the applicability area of the industrial products. The difference between the manufacturing recipes of the variants 2 and 3 consists both in the glass waste nature and in the weight proportions of glycerol, sodium silicate and water used in the two cases.

Variant 2 exclusively used flat glass waste, while variant 3 used container glass waste composed of colorless, green and amber glass in the 50/20/30 ratio, considered as real for the recycled glass in Romania. Referring to the liquid components proportions in mixing with the raw material, in variant 2 they were used 1.3 wt.% glycerol and 5.9 wt.% sodium silicate, in a 1:4.5 ratio and in variant 3, 1.0 wt.% glycerol and 8.0 wt.% sodium silicate, in a 1:8 ratio. The water, that has a role in the glycerol dissolving in the liquid solution before mixing with the glass powder, was used in proportions of 9.1 wt.% and 8.0 wt.%, respectively.

If, regarding the chemical composition of the glass waste used in the two variants there are no significant differences that influence the quality of the final product, certain discussion can be made referring to the proportions of the liquid components of the mixture. Thus, the weight proportion of glycerol lower in variant 3 (1 wt.%) compared to variant 2 (1.3 wt.%) could have influenced the pore fineness in the first case. On the other hand, the higher proportion of sodium silicate of variant 3 (8 wt.%) could have influenced to obtain a higher mechanical strength (5.9 MPa) compared to variant 2 (5.3 MPa), being known in the literature [22] its role in improving the mechanical properties.

Therefore, based on the comparative analysis of the characteristics of the foam glass gravel samples experimentally produced in the microwave field, the best sample was considered the sample 3, whose manufacturing recipe comprised 1 wt.% glycerol as a foaming agent, 8 wt.% sodium silicate as an enveloping material and 8 wt.% water. The product had apparent density of 0.24 g/cm^3, porosity of 89.1%, thermal conductivity of 0.063 W/m·K, compressive strength of 5.9 MPa and water absorption of 0.6%. The foamed material had a homogeneous porous structure with very low pore size between 0.3-0.6 mm. The manufacturing process required a maximum heating temperature of 823 ºC reached in 42 min with an average heating rate of 19.1 ºC/min. The specific energy consumption of the process was very low (0.88 kWh/kg) [26]. The data provided by the literature regarding this functional parameter are very general and inconclusive [2]. A specific consumption around 0.85-0.90 kWh/kg could be attributed to the industrial processes of manufacturing foam glass gravel by conventional techniques. Therefore, the consumption value achieved under experimental conditions on a very low power oven and with discontinuous operation was within the limits of the industrially obtained values. According to the literature [16], a microwave equipment of high-power equivalent to the industrial conditions of production could reach an energy efficiency of up to 25% compared to the experimental oven of only 0.8 kW. Therefore, in energy terms, the unconventional heating technique experimentally applied could be significantly more efficient than the conventional techniques currently used in industry.

4. CONCLUSIONS

The research, whose results are presented in the paper, carried out a comprehensive comparative analysis of several types of foam glass gravel experimentally produced by heating in microwave field using different manufacturing recipes.

The originality of the research consists in the use of an advanced technique of unconventional heating of glass waste-based raw material for the manufacture of foam glass gravel, unlike the conventional techniques currently used in the world industrial production.

Two manufacturing variants using glycerol as a liquid foaming agent and two variants using solid foaming agents (calcium carbonate and silicon carbide, respectively) were compared. The main difference between the variants using glycerol consisted of the type of glass waste used: flat glass waste and container glass waste (colorless, green and amber), respectively.

The sample made with silicon carbide as well as the one made with calcium carbonate, borax and sodium silicate had very high mechanical strength (7.4-7.5 MPa), but their apparent density was too high (0.35 g/cm^3 and especially 0.62 g/cm^3) for a quality foam glass gravel. The samples made with glycerol had a light weight (0.24 g/cm^3) and the compressive strength was within the acceptable range for this type of cellular glass (5.3-5.9 MPa).

Following the comparative analysis it resulted that the best sample of foam glass gravel was that made of 83 wt.% container glass waste, 1 wt.% glycerol, 8 wt.% sodium silicate and 8 wt.% water by heating to 823 ºC with a low energy consumption of 0.88 kWh/kg. The sample characteristics were: apparent density of 0.24 g/cm^3, porosity of 89.1% with pore size between 0.3-0.6 mm, thermal conductivity of 0.063 W/m·K, compressive strength of 5.9 MPa and water absorption of 0.6%.

The specific energy consumption of the experimental process was 0.88 kWh/kg, which is almost similar to that achieved in the industrial production of foam glass gravel, could theoretically be reduced by up to 25% by applying the unconventional heating technique on an industrial microwave equipment.

REFERENCES

[1] Scarinci, G., Brusatin, G., Bernardo, E., Cellular ceramics: structure, manufacturing, properties and applications, Scheffler, M., Colombo, P. (eds.), Wiley-VCH Verlag GmbH and KGaA, Weinheim, 2005, p. 158-176.
[2] Hurley, J., Glass-research and development, Final report, A UK market survey for foam glass, The Waste and Resources Action Programme Publication, Banbury, Oxon, 2003.
[3] Hibbert, M., Understanding the production and use of foam glass gravel across Europe and opportunities in the UK, Final report, Chartered Institution of Wastes Management, Northampton, July 2016.
[4] www.geocell-schaumglas.eu/en/ (15.12.2019).
[5] www.archiexpo.com>soc>misapor-78710 (10.03.2020).
[6] www.misapor.ch/en/company (16.02.2020).
[7] www.archiexpo.com/soc/misapor-78710.html (10.05.2019).
[8] www.Glapor_cellular_glass_gravel (15.09.2019).
[9] www.glapor.de/en/produkte/cellular_glass_gravel (12.01.2020).
[10] www.glamaco.com/products/foam-glass-gravel (08.02.2020).
[11] www.glamaco.com/backend/wp-content/uploads/2014/02/Foam-Glass.pdf (15.03.2020).
[12] www.refaglass.cz/en/ (02.03.2020).

[13] www.environdec.com/Detail/?Epd=13145 (20.03.2020).
[14] www.glasopor.no/om-produktet/lav-vekt/ (20.03.2020).
[15] www.foamit.fi/en/foamit-products/ (20.03.2020).
[16] Kharissova, O.V., Kharissov, B.I., Ruiz Valdés, J.J., Review: The use of microwave irradiation in the processing of glasses and their composites, Industrial and Engineering Chemistry Research, vol. 49, no. 4, 2010, p. 1457-1466.
[17] Knox, M., Copley, G., Use of microwave radiation for the processing of glass, Glass Technology, vol. 38, no. 3, 1997, p. 91-96.
[18] Paunescu, L., Axinte, S.M., Grigoras, B.T., Dragoescu, M.F., Fiti, A., Testing the use of microwave energy to produce foam glass, European Journal of Engineering Technology, vol. 5, no. 4, 2017, p. 8 – 17.
[19] Karandashova, N.S., Goltsman, B.M., Yatsenko, E.A., Analysis of influence of foaming mixture components on structure and properties of foam glass, IOP Conference Series: Materials Science and Engineering, vol. 262, 2017, https://iopscience.iop.org>article>262 (16.01.2020).
[20] Brusatin, G., Bernardo, E., Scarinci, G., Production of foam glass, Proceedings of the International Conference Sustainable Waste Management and Recycling: Glass Waste, Kingston University of London, September 14-15, 2004, p. 68-82.
[21] Eidukyavichus, K.K., Matseikene, V.R., Balkyavichus, V.V., Shpokauscas, A.A., Use of cullet of different chemical compositions in foam glass production, Glass and Ceramics, vol. 61, no. 3-4, 2004, p. 77-80.
[22] Ayadi, A., Stiti, N., Benhaoua, E., Boumchedda, K., Lerari, Y., Elaboration and characterization of foam glass based on cullet with addition of soluble silicates, Proceedings of the International Conference on Advances in Materials and Processing Technologies, vol. 1315, no. 1, Paris, October 26-27, 2010.
[23] www.docplayer.net/21731890-Manual-of-weighing-applications-part-1-density.html (15.12.2019).
[24] Anovitz, L.M., Cole, D.R., Characterization and analysis of porosity and pore structures, Reviews in Mineralogy and Geochemistry, vol. 80, 2005, p. 61-164.
[25] Bianchi-Janetti, M., Plaz, T., Ochs, F., Klesnil, O., Feist, W., Thermal conductivity of foam glass gravel: a comparison between experimental data and numerical results, Energy Procedia, vol. 78, 2015, p. 3258-3268.
[26] Paunescu, L., Dragoescu, M.F., Axinte, S.M., High mechanical strength porous material used as a foam glass gravel experimentally manufactured from glass waste by an unconventional technique, Construction (in process of publication).