CELLULAR GRAVEL OF SLAG AND COAL ASH MADE BY AN UNCONVENTIONAL HEAT TREATMENT METHOD

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***ABSTRACT**: The paper presents authors' original contributions in the experimental manufacturing process of cellular gravels with good thermal insulation properties (bulk density between 0.29-0.38 g/cm^3 and thermal conductivity between 0.092-0.105 W/m·K) and very high compressive strength (between 12.6-14.2 MPa). Unlike all heat treatment processes used in the world in this domain, the heating technique is unconventional based on the direct microwave irradiation. As a result, the heating rate reached very high values (32.3-40.1 ºC/min) without affecting the structural homogeneity of products. Another originality of the work is the use as raw material of other silicate waste (blast furnace slag and coal fly ash) except glass waste, which seemed technologically irreplaceable.*

***KEYWORDS**: cellular gravel, direct microwave heating, coal fly ash, blast furnace slag*

1 INTRODUCTION

After 1985, the cellular glass gravel made from recycled glass waste became a product of great interest for construction works such as: insulation under the ceiling, thermal insulation of terraces and balconies, flat roof insulation for green areas, load bearing thermal insulation for under foundation slabs and perimeter insulation of building. Also, cellular glass gravel is used in road and railway construction, bridge abutments and retaining walls, insulation of underground pipelines and storage tanks, drainage, sports fields, etc. (Product Declaration, 2017; Product Declaration, 2014; Geocell, 2017). Above 600,000 m^3 per year are produced in Europe, in Germany, Switzerland, Austria and the Nordic countries (Hibbert, 2016). This cellular material uses recycled glass waste mainly from post-consumer drinking bottle or flat glass waste from demolition or renovation of buildings and a foaming agent that can be coal, black carbon, graphite, glycerol, calcium carbonate, sodium carbonate, dolomite, silicon carbide, silicon nitride, etc. (Scarinci et al., 2005). The characteristics of cellular glass are remarkable. Except for the thermal insulation properties (light weight, high porosity and low thermal conductivity), it have also a good resistance to fire, humidity, frost, corrosion, resistance to rodent and insect aggression, very high durability, physical and chemical stability, lack of toxicity, etc. (Cellular glass, 2016).

Not only the glass waste has properties that allow the manufacture of cellular materials, but also other industrial silicate waste or by-products (metallurgical or thermal power station slag, coal fly ash, waste incinerator ash, mud from zinc hydrometallurgy, etc.) are suitable for this purpose, but generally, as partial substitutes for glass waste (Scarinci et al,. 2005; Rawlings et al., 2006).

In many cases, silicates-based industrial waste or by-products can contain contaminants for the environmental. The vitrification methods of this waste, i.e. transforming the silicates into an amorphous mass by heating to melting temperature followed by solidification by cooling, practically include glass-ceramic manufacturing methods. The use of a suitable foaming agent in the predominant mixture of silicates produces cellular glass-ceramics. The new products obtained by vitrification, generally regardless of the contaminant level of the waste, have numerous applications, especially in the construction sector.

Metallurgical slag and especially blast furnace slag is an industrial waste found in large quantities in the dumps of metallurgical plants around the world. A major disadvantage in the vitrification process could be an iron-rich waste, which can negatively influence the porosity of cellular glass materials and also catalytic, electrical, magnetic and optical properties, while a silica-rich waste favors

the obtaining of an amorphous glassy structure (Rincón et al., 2006).

The blast furnace slag preferably rich in titanium oxide (Sun & Wang, 2017), granulated by water cooling, together with glass waste, borax (optimally 8 %) as a fluxing agent and calcium carbonate ($CaCO_3$) as a foaming agent, sintered at 900 ºC for 30 min, had uniform porous structure, bulk density of 0.82 g/cm^3 and a very high compressive strength (up to 25 MPa).

According to (Wang et al., 2018), increasing the proportion of $CaCO_3$ as a foaming agent led to intensifying the foaming process. As a consequence, the bulk density and the compressive strength increased, while porosity and water absorption were reduced. For proportions of the foaming agent between 5-7 %, the bulk density had values in the range 0.79-0.82 g/cm^3, the porosity was between 73.1-75.3 %, the compressive strength was between 13.1-13.8 MPa and the water absorption was below 3.7 %.

When blast furnace slag is not rich in titanium (TiO_2) (Ding et al., 2015), this oxide together with zirconia (ZrO_2) and calcium fluoride (CaF_2) have been added as nucleating agents in the slag and glass waste mixture. Other material additives were borax, sodium phosphate ($Na_3PO_4 \cdot 12H_2O$) and $CaCO_3$ (about 6 %) as a foaming agent. Glass-ceramic foams with 50 % blast furnace slag led to excellent comprehensive properties (bulk density of 0.79 g/cm^3, water absorption of 2.7 % and compressive strength of 14.3 MPa.

A manufacturing recipe approximately similar to that presented in (Ding et al., 2015), containing blast furnace slag and glass waste, borax as a fluxing agent, TiO_2 as a nucleating agent, $Na_3PO_4 \cdot 12H_2O$ as a stabilizer, $CaCO_3$ as a foaming agent and water as a binder, was used by a team from the Romanian company Daily Sourcing & Research which includes authors of current work. Unlike the conventional heating method used in (Ding et al., 2015) and in all the works cited above, the paper (Grigoras et al., 2020) applied an own unconventional microwave heating technique. The pressed powder mixture was sintered at temperatures between 900-905 ºC using slag/glass ratios between 40/60 and 50/50. The heating rate had high values (20.11-21.20 ºC/min) and the specific energy consumption was very low (0.90-0.95 kWh/kg). The optimal variant included the 40/60 ratio of the two raw material types, 7.8 % borax, 5 % TiO_2, 3 % $Na_3PO_4 \cdot 12H_2O$, 6.5 % $CaCO_3$ and 8 % water addition and the foamed sample features were: apparent density of 0.82 g/cm^3, porosity of 75.9 %, thermal conductivity of 0.135 W/m·K, compressive strength of 14.1 MPa and water absorption of 3.4 %.

The coal fly ash is an industrial by-product of thermal power stations as a result of coal burning. Worldwide, it has a variable chemical composition, in which silica (SiO_2) has values between 42.8-66.1 % and alumina (Al_2O_3) in the range 11.4-29.6 % (Wu, 2006), being an aluminosilicate material. According to the literature, the use of coal fly ash in very high weight proportions led to the manufacture of cellular glass-ceramics with very high mechanical strength.

A conventional heating manufacturing method of a cellular glass-ceramic with high mechanical strength was applied by the authors of the paper (Ma et al., 2018). According to the information in literature, an extremely high weight proportion of coal fly ash (between 83-95%) as raw material, 5 % sodium oxide (Na_2O) as a fluxing agent and $CaCO_3$ (between 3-9%) as a foaming agent were mixed and sintered at 1150 ºC. The physical and mechanical characteristics of the products were: bulk density between 1.55-1.59 g/cm^3, porosity between 16.8-19.9%, bending strength in the range 43.4-109.6 MPa. The uniformity of the cells size was generally good, the product made with 6% $CaCO_3$ having the highest microstructural homogeneity (between 0.1-0.4 mm).

The paper (Paunescu et al., 2020) presents a high mechanical strength cellular glass-ceramic produced by the direct microwave heating technique at 853 ºC of a very high weight ratio of coal fly ash (82%), 5 % $CaCO_3$ as a foaming agent, 13 % sodium carbonate (Na_2CO_3) as a fluxing agent and 10% water addition as a binder. The direct microwave heating technique allowed to reach a very high heating rate (32 ºC/min) and shortening the heating time without affecting the microstructure of the heat-treated sample. The specific energy consumption was significantly reduced compared to conventional heating methods, reaching 0.72 kWh/kg. The main features of the optimal product were: apparent density of 1.44 g/cm^3, porosity of 26.2 %, thermal conductivity of 0.281 W/m·K, compressive strength of 41.3 MPa and water absorption of 0.5 %. The work was performed by Daily Sourcing & Research Company (Grigoras et al., 2020), using the same unconventional microwave heating technique. Given the features of the cellular glass-ceramic (very high compressive strength, acceptable porosity and thermal conductivity, very low water permeability, fireproof, chemical and physical stability, no-toxicity, high durability, etc.), the application field of this material type may include road and railway constructions, bridge abutments and retaining walls,

foundations, drainages, sports grounds and other construction types that require high mechanical stress.

Jarosite, an iron-rich waste resulting from zinc hydrometallurgy process, was used (in proportions between 40-70%) together with granite scraps and mud (between 20-40%) and glass waste (between 10-40%) to produces glass-ceramic materials by sintering at 1400-1450 ºC (Pelino, 2000). The final product (with 40-55% crystalline volume fraction and high compressive strength up to 10 MPa) was achieved in the form of paving tiles, wall panels and glass fibers for building construction.

Oil shale ash waste resulted from thermal power station constituted the main raw material for manufacturing glass-ceramics (Gorokhovski et al., 2002). The oil shale-based glass, with small additions of glass and sand waste, was melted at 1400 ºC and cooled with water in the form of fritted glass. $CaCO_3$ (optimal 5%) was the foaming agent. Sodium metasilicate (Na_2SiO_3) and oil shale waste were added in the ratio 1/5 to the total glass waste. The temperature of the thermal process was 900 ºC and the final product had good thermal insulation properties and relative normal mechanical strength (flexural strength of 3 MPa).

The current work aimed to find a technical solution for the manufacture of a glass-ceramic that combines very high mechanical strength with bulk density and thermal conductivity at much lower levels compared to the products previously made and presented above. The content of the paper is original from two different points of view. On the one hand, the starting raw material does not contain glass waste, but only silicate waste usually used in lower proportions as partial glass substitutes (blast furnace slag and coal fly ash). On the other hand, the direct microwave heating method of material mixture is original. Due to the sufficiently high content of silica and alumina, especially the coal ash, the mixture becomes an aluminosilicate material. Previous own experiments (Paunescu et al., 2019) have shown that such a ceramic material is suitable for the direct microwave heating (unlike to that with high glass content), without the extremely high heating rate affecting its microstructural configuration.

2 MATERIALS AND METHODS

2.1 Materials

As mentioned above, the main wastes used as raw material in the current experiment were: blast furnace slag granulated by water quenching from the metallurgical plant ArcelorMittal Galati (Romania) between 44-53 % and coal fly ash, a by-product resulted by coal burning in the Paroseni thermal power station between 37-46 %. The solid mixture was supplemented with sodium borate (borax), an excellent fluxing agent due to the high Na_2O content and zirconia (ZrO_2) as a nucleating agent. The weight ratios of the two agents were kept constant for all tested variants: 8 % for borax and 2 % for zirconia.

The blast furnace slag granulated by fast water cooling of the molten mass initially had a grain size below 2.5 mm. Its final granulation was reduced below 130 μm by grinding in a ball mill and sieving.

The coal fly ash purchased from Paroseni power plant had the grain size below 200 μm being need his mechanical processing in the ball mill for reducing the maximum size up to 100 μm.

Table 1 presents the oxide composition of the two main components of raw material used in the experiment.

Table 1. Oxide composition of raw material

Oxide composition	Blast furnace slag (wt. %)	Coal fly ash (wt. %)
SiO_2	36	46.5
Al_2O_3	7-9	23.7
CaO	41	7.9
MgO	7-8	3.2
Na_2O	-	6.0
K_2O	-	4.1
Fe_2O_3	0.5-1.5	8.6
FeO	0.5-0.8	-
S	0.8	-
MnO	0.2-1	-

The oxide composition of blast furnace slag was taken from the work (Cioroi & Nistor-Cristea, 2007) and the coal fly ash composition was offer by the Paroseni power plant.

It should be mentioned that the blast furnace slag purchased from ArcelorMittal Galati (Romania) has a sufficiently low iron content (such as Fe_2O_3 and FeO), so that the negative influence on the porosity of cellular glass-ceramic is not exhibited according to Rincón et al., 2006.

One of the optimal nucleating agents in Li_2O-Al_2O_3-SiO_2 glass-ceramic system is zirconia (ZrO_2) with a monolithic crystal structure at room temperature, whose granulation is very fine (below 15 μm). Due to the use of this agent, the crystallization energy is generated and glass ceramic with low cell size and high mechanical properties can be obtained (Hu et al., 2009).

The borax presence in the starting mixture has a double role. On the one hand, the presence of Na_2O in a high proportion in the composition of borax provides it with important flux properties. Consequently, borax was adopted as a fluxing agent. On the other hand, the boron in the composition of borax has the ability to increase the mechanical strength of the ceramic product manufactured by adding this material.

The adopted foaming agent was glycerol ($C_3H_8O_3$), a liquid carbonaceous agent. Commonly, glycerol is associated with water soluble sodium metasilicate (Na_2SiO_3) called also "water glass", which has the role of slowing down the premature decomposition of glycerol and thus to promote sintering the solid mixture and pure water as a binder. The liquid mixture was separately prepared, the weight ratios of glycerol, water glass and pure water being 1/ 8/ 5. The use of an aqueous foaming agent in the manufacture of cellular glass-ceramic allows an intimate mixture to be achieved through an advanced degree of aqueous solution penetration through the fine solid particles and consequently favors the foaming process, reduces the cells size and decreases the material density.

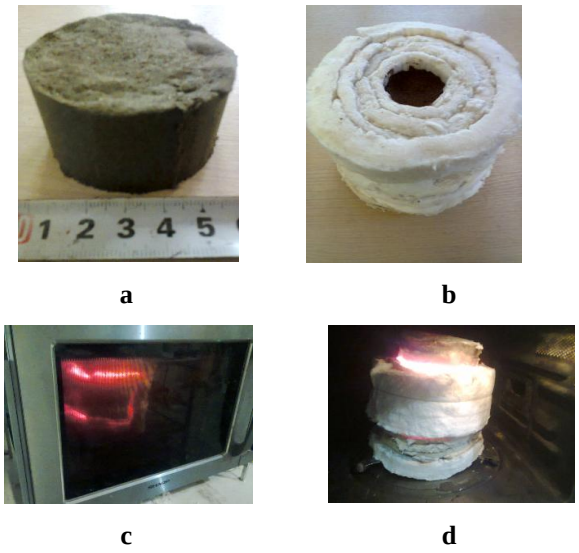
The thermal decomposition of glycerol is a very complex process. Several compounds between carbon dioxide and pure carbon as well as hydroxyl compounds are formed by decomposition in the oxidizing atmosphere of the oven, according to (Karandashova et al., 2017; Zhang et al., 2021).

2.2 Methods

The adopted method for the experimental manufacture of a cellular glass-ceramic was the following. The mixture of the solid raw material and the addition of the aqueous solution of glycerol-water glass-pure water (prepared separately) was made in a metal mold with an inner diameter of 50 mm, pressed axially at about 10 MPa and then removed from the mold. Prior to introduction into the 800 W-microwave oven of the type commonly used in the household, but constructively adapted for high temperature operation (above 1000 ºC), the pressed powder mixture was thermally protected with several ceramic fiber mattresses resistant to 1200 ºC. A thick bed of ceramic fiber mattresses was placed at the base of the oven, on which the thermally protected pressed mixture was stored. The upper part of the ceramic package containing the material was covered with a ceramic fiber lid provided with a central hole of 25 mm for viewing with a radiation pyrometer mounted above the oven of the upper surface of the heated sample. Also, the upper metal wall of the oven had a central axial hole of 25 mm for viewing with the pyrometer.

The direct microwave heating, as an unconventional heating technique, has some peculiarities that make it completely different compared to conventional techniques commonly used in similar processes. The main peculiarity is the initiation of the heating process in the core of the microwave irradiated material followed by the volumetrically transmission of heat throughout the mass of the material from inside to outside (Jones et al., 2002). Therefore, the outer surface of the heated sample must be thermally very efficiently protected. Thus, the metal wall of the oven no longer needs to be protected. Another important peculiarity of the direct microwave heating is the selectivity of the thermal process (Kitchen et al., 2014). Thus, only the material subjected to irradiation absorbs electromagnetic waves that are converted into heat, not other massive components of the oven (usually, refractory ceramic materials) in which the process takes place and which are not microwave susceptible materials. The two peculiarities mentioned above have a remarkable effect in terms of energy on the heating process. This is significantly faster and more economical compared to conventional methods which is also noted in (Aggregates, 2016). What has been experimentally observed by the authors' team is that the very high heating rate does not affect the microstructural characteristics of the foamed product.

The main details of preparing the raw material (a), its thermal protection with ceramic fiber mattresses (b) and unconventional heating into the microwave oven (c and d) are shown in Figure 1.



a **b**

c **d**

Fig. 1 Details of the manufacturing process

a – pressed powder raw material; b – ceramic fiber thermal protection of raw material; c – 800 W-microwave oven during the heating process; d – protected sample image inside the oven at the end of the process.

2.3 Adoption of manufacturing recipe

The manufacturing recipe adopted by the authors consisted of a solid mixture including blast furnace slag, coal fly ash, borax and zirconia in which the variables were only the slag (between 44-53 %) and the ash (between 37-46 %). The liquid mixture was kept constant during the experiment. Table 2 presents the distribution of the two mixture types in four variants.

Table 2. Distribution of solid and liquid mixture

Mixture type	**Variant**			
	1	**2**	**3**	**4**
				Solid mixture
Blast furnace slag	44	47	50	53
Coal fly ash	46	43	40	37
Borax	8	8	8	8
Zirconia	2	2	2	2
				Liquid mixture
Glycerol	1	1	1	1
Water glass	8	8	8	8
Pure water	5	5	5	5

3 RESULTS AND DISCUSSION

3.1 Results

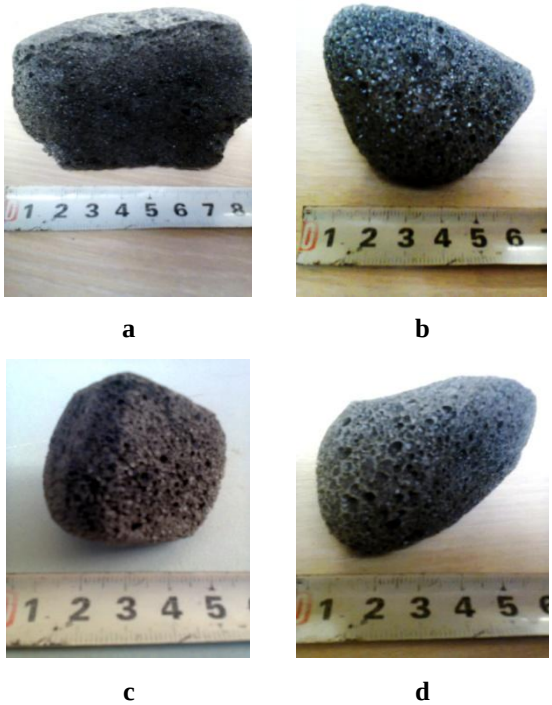
A relatively constant amount of wet raw material (235.4-236.1 g) was pressed and inserted into the microwave oven to be sintered for foaming. The main functional parameters of this process are presented in Table 3.

Table 3. Parameters of the direct microwave heating process

Parameter	Variant			
	1	2	3	4
Wet raw material/cellular gravel amount (g)	235.4/ 188.3	236.0/ 191.2	235.8/ 189.8	236.1/ 191.1
Sintering temperature (ºC)	902	908	915	924
Heating time (min)	22	23	25	28
Average heating rate (ºC/min)	40.1	38.6	35.0	32.3
Average cooling rate (ºC/min)	7.0	7.1	6.8	7.1
Specific energy consumption (kWh/kg)	1.22	1.25	1.37	1.53

The heating time required to reach the foaming temperature of 902-924 ºC was very short (between 22-28 min). The calculation of the heating rate led to extremely high values (between 32.3-40.1 ºC/min), much higher compared to the rates corresponding to the conventional heating processes (around 10 ºC/min). By adopting higher average cooling values (around 8 ºC/min) internal stresses are created in the structure of material, which facilitates the detachment of lumps with adequate dimensions (40-70 mm).

Images of cellular gravel lumps obtained by heat treatment of the waste mixture in the four heating variants are shown in Figure 2.



a **b**

c **d**

Fig. 2. Images of the cellular gravel lumps

a – variant 1; b – variant 2; c – variant 3;

d – variant 4.

According to Figure 2, the appearance of the four cellular gravel samples is slightly different, their granulation varying to a relatively small extent. However, a slight but noticeable increase of the sample cell size can be observed when the slag/ash ratio increases from 44/46 to 53/37 (i.e. from variant 1 to variant 4).

In order to have a clear perspective of the influence of changing this proportion on the

physical, thermal, mechanical and microstructural characteristics of the expanded products, it was necessary to identify the value of these characteristics. The bulk density was measured by dividing the mass of the fully loaded lumps into a cylindrical vessel at its known volume (Aggregates, 2016). The porosity was calculated by the method of comparing the compact material density (true density) and the porous material density (Anovitz & Cole, 2005). The thermal conductivity was measured by the heat-flow meter method (ASTM E1225-04) and the compressive strength was determined using the TA.XTplus Texture Analyzer (ASTM C552-17). The water absorption was determined by the water immersion method (ASTM D570) and the microstructural features of the cellular gravel were investigated with a Smartphone Digital Microscope ASONA 100X Zoom type.

The results of identifying the cellular gravel characteristics are presented in Table 4.

Table 4. Main characteristics of the cellular gravel samples

Characteristics	Variant			
	1	2	3	4
Bulk density (g/cm^3)	0.38	0.35	0.31	0.27
Porosity (%)	71.9	73.3	75.2	77.1
Thermal conductivity (W/m·K)	0.105	0.100	0.096	0.092
Compressive strength (MPa)	12.9	12.6	12.8	14.2
Water absorption (vol. %)	3.4	3.1	3.1	2.9
Cells size (mm)	0.1-0.4	0.3-0.5	0.3-0.8	0.8-1.0

The effect of easily penetrating the fine spaces between the solid particles of the mixture by the aqueous solution of glycerol and sodium silicate can be observed on the relatively low values of bulk density (between 0.27-0.38 g/cm^3) compared to those of expanded products with solid foaming agents (0.75-0.95 g/cm^3 and even higher). Also, the thermal conductivity has relatively low values, that do not exceed 0.105 W/m·K thus contributing to ensuring the thermal insulation character of this foamed mixture of blast furnace slag and coal fly ash. The high comprehensive properties of the material are given by the compressive strength, which reaches very high values for a ceramic product (between 12.6-14.2 MPa) and the low water absorption (below 3.4 vol.%). The liquid foaming agent used in the experiment also contributed to a very fine and homogeneous microstructural configuration with a uniform cell distribution. The cellular structure is generally closed, with no tendency to communicate between cells, according to Figure 3.

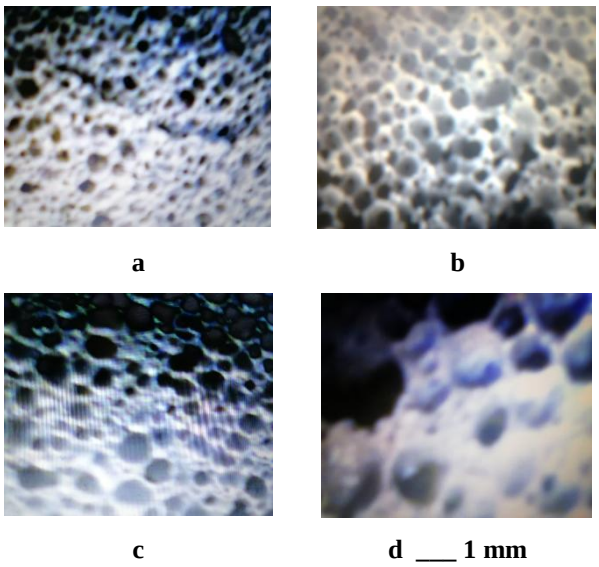


Fig. 3. Microstructural characteristics of the cellular gravel lumps

a – variant 1 expanded at 902 ºC; b – variant 2 expanded at 908 ºC; c – variant 3 expanded at 915 ºC; d – variant 4 expanded at 924 ºC.

3.2 Discussion

Currently, the manufacture of porous thermal insulation materials, dense and with a high mechanical strength is made industrially from silicate waste, but the predominant waste is recycled glass. Ceramic gravels in the form of lumps with approximate dimensions between 10-70 mm are used in various types of construction works as thermal insulation filling materials subjected to mechanical stress and often in difficult environmental conditions. The parental characteristics of glass as a raw material (resistance to various factors, durability, physical and chemical stability, non-toxic, etc.) are considered appropriate for this type of product.

Experimental research performed in the world investigates the influence of the combination of glass waste as a basic raw material with other waste types, aiming to obtain materials with thermal insulation properties and simultaneously with very high mechanical strength.

The originality of the current work is the experimental manufacture of ceramic gravel using exclusively other silicate or aluminosilicate waste excluding glass waste. Also, a high degree of the paper originality is applying the direct microwave heating technique, a faster and more economical unconventional method, which is not used industrially or on a small experimental scale than in processes of drying and low temperature heating.

These technological changes in the manufacturing process of cellular gravel (using slag and coal ash wastes and applying the direct microwave irradiation of raw material) did not influence the physical, thermal, mechanical and microstructural properties of the final product. Furthermore, the energy efficiency of the unconventional process is superior to conventional processes.

It is necessary to prove in industrial conditions the conclusions resulting on a small scale and this objective is the authors' concern for future stages.

4 CONCLUDING REMARKS

The work objective was testing the use in the process of manufacturing the cellular gravel of a starting powder mixture including as raw material a silicate waste (granulated blast furnace slag) and an industrial by-product (coal fly ash), except glass waste always used in similar processes. An aqueous solution of glycerol together with sodium silicate (water glass) was adopted as a foaming agent, because it easily penetrates the fine particles of solid waste. Along with the exception of glass waste, another originality of the work was applying the unconventional method of direct microwave heating, unlike the conventional methods used in the world. The results were excellent, being manufactured products with bulk density between 0.27-0.38 g/cm^3, thermal conductivity between 0.092-0.105 W/m·K and very high value of compressive strength (12.6-14.2 MPa) suitable for using as cellular gravels in construction works. The excessively high heating rate (up to 40.1 ºC/min) did not affect the structural homogeneity of the material.

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