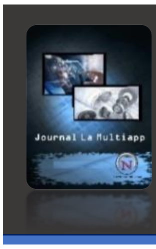

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Experimental Use of Microwaves in the High Temperature Foaming Process of Glass Waste to Manufacture Heat Insulating Materials in Building Construction

Lucian Paunescu[1], Sorin Mircea Axinte [1,2],
Marius Florin Dragoescu[1], Felicia Cosmulescu[3]

[1] Daily Sourcing & Research SRL Bucharest, Romania
[2] University "Politehnica" of Bucharest, Department of Applied Chemistry and Materials Science, Bucharest, Romania
[3] Cosfel Actual SRL Bucharest, Romania

Corresponding Author: Lucian Paunescu
Email: lucianpaunescu16@gmail.com

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Abstract

The aim of the paper was the experimental manufacture of cellular glass from glass waste and coal ash as raw material and silicon carbide as a foaming agent, using the unconventional microwave heating technique. This heating technique, although known since the last century and recognized worldwide as fast and economical, is not yet industrially applied in high temperature thermal processes. The cellular glass manufacturing process requires high temperatures and the use of microwaves in this process is the originality of the work. The experiments aimed at producing thermal insulating materials with high porosity and low thermal conductivity for building construction similar in terms of quality to those manufactured industrially by conventional techniques, but with lower energy consumption. The obtained samples had adequate characteristics (apparent density 0.22-0.32 g/cm3, porosity 85.5-90.0%, thermal conductivity 0.043-0.060 W/m•K, compressive strength 1.23-1.34 MPa), and the specific energy consumption was low (0.84-0.89 kWh/kg). Theoretically, given the use of microwave equipment on an industrial scale, this consumption comparable in value to that industrially achieved by conventional techniques could decrease by up to 25%.

Introduction

A large amount of glass waste, especially post-consumer container glass, is available worldwide, and the annual rate of its generation at least in recent decades is very high. The glass industry uses part of this reserve as a raw material for the manufacture of the new glass. It should be noted that the cost of the process of sorting colored glass from the waste batch as an important stage of the industrial glass manufacturing process is quite high. This is the main reason why it is much more profitable to use glass waste as the predominant raw material for the manufacture of cellular glass as a replacement for existing construction materials. According to the literature, the industrial production of 1 kg of new glass requires an energy consumption of about 4500 kJ, while the industrial manufacture (by conventional methods of heat treatment) of 1 kg of cellular glass requires only 500 kJ or 0.56 kWh (Da Silva et al., 2016; Energocell, 2014).

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In principle, the manufacture of cellular glass involves the expansion of the very finely ground glass-based raw material, which has incorporated in its mass, and homogeneously mixed a relatively small amount of foaming agent (black carbon, coal dust, graphite, calcium carbonate, silicon carbide, glycerine, etc.) capable of releasing a gas at high temperature (750-1100 ºC) (Scarinci et al., 2005). It is essential that the beginning of the gas release to take place at a temperature at which the raw material powder mixture has an adequate viscosity due to the thermal softening, so that the gas resulting from a decomposition or oxidation process to spread evenly in the material mass, but to cannot leave it, remaining blocked. By cooling, the gas bubbles contribute to the formation of a homogeneous porous structure (Scarinci et al., 2005; Paunescu et al., 2017). Thus produced, the cellular glass simultaneously includes uniquely cumulated properties (low density, low thermal conductivity, high enough compressive strength, fire resistance, water and steam impermeability, frost-thaw resistance, resistance to the aggression of rodents, insects, bacteria or acids) (Scarinci et al., 2005). Due to these unique characteristics, the cellular glass is a viable alternative to currently using polystyrene as a thermal insulation material in the building construction. According to the literature (Energocell, 2014), the primary energy required for manufacturing the cellular glass is about 140 kWh/m^3, i.e. between 0.80-0.93 kWh/kg, taking into account the density values of this product between 150-175 kg/m^3, being much lowest compared to the primary energy required to produce polystyrene as a heat insulating material (about 1400 kWh/m^3).

Depending on the nature of the foaming agent, but also on other types of silicate waste and various mineral additives added to the manufacturing recipe, the cellular glass can have different physical and mechanical characteristics, and can be used both as a light insulating material with acceptable compressive strength up to 2.75 MPa (Foamglas, 2013) and as a much denser material with relatively high compressive strength (up to 6 MPa or even more) used as foundation material in lightweight construction, road and railway construction, airport runways, drainage, sports fields, etc. (Technopor, 2012).

It should be noted that all types of cellular glass commonly manufactured in industry as well as those made in the laboratory as variants of manufacturing recipes, are produced by conventional heat treatment techniques (electrical resistances or burners using fossil fuels).

In recent years, the concern of the authors of the current paper has been focused on the use in the manufacturing process of cellular glass of an unconventional heating technique: the microwave heating. Although known since the middle of the last century, and despite the fact that it is recognized in the world as a fast, "clean" and economical heating technique, the application of microwaves has been done to a small extent, more in the household for food preparation and industrially only in drying or heating at low temperature processes. In the last 2-3 decades, it was experimentally found that several material types (organics, ceramics, polymers, metals, glass, etc.) are suitable for the microwave heat treatment, but the results are still on experimental stages (Kharissova et al., 2010).

Regarding the use of microwaves in the cellular glass manufacture, a market study (Hurley, 2003) concluded in 2003 that the transformation of existing tunnel ovens in the UK into microwave ovens would not be efficient because, given that the glass contains mainly silica and alumina which there are no microwave susceptible materials at low temperatures below 500 ºC (Knox & Copley, 1997), it would be necessary to equip ovens with conventional heating installations in areas below 500 ºC, and with microwave generators in the hot areas (over 500 ºC).

Since 2016, the Romanian company Daily Sourcing & Research SRL Bucharest has successfully carried out a series of experiments aiming to obtain cellular glass from glass waste, and these tests have shown that the glass waste can be microwave heated quickly and efficiently from room temperature (Paunescu et al., 2017). The reason, theoretically confirmed by other researchers (Jones et al., 2002; Kolberg & Roemer, 2001), is that in the commercial glass composition used as a raw material there are inherently certain contaminants (Fe_2O_3, Cr_2O_3, etc.) with high microwave susceptibility at room temperature. The current work contains results of the experimental manufacture of a light porous cellular glass type with thermal insulating properties using the microwave energy.

Methods

Unlike other known types of conventional energy sources, the electromagnetic wave (radiated in the frequency range between 300 MHz-300 GHz) should not be considered as a form of energy. The intense thermal energy developed in the irradiated material under the influence of a microwave field results from the conversion of these waves into heat through direct contact with the material. The condition required for this thermal process to occur is that the material subjected to heating be a microwave susceptible material. In the case of a powder mixture, at least one of the components of the mixture should be microwave susceptible (Jones et al., 2002).

The use of microwaves in high temperature heating processes of solid materials is advantageous due to some special peculiarities. Thus, the heating is selectively performed, only the material subjected to the process being heated, without the need to heat the other massive components of the oven as in the case of conventional processes (Jones et al., 2002). Also, the heating due to the microwave irradiation is initiated in the core of the material, where the highest temperature is reached, the heat being then propagated to the peripheral areas of the material. Because the material itself generates heat, the process takes place volumetrically and can be very fast (Kitchen et al., 2014). Also, the presence in important proportion in the composition of the glass, as the main raw material, of the oxides of the alkali metals (Na_2O, K_2O) favors the microwave absorption process (Kolberg & Roemer, 2001). Due to these microwave heating peculiarities, the heating rate is higher, the specific energy consumption is lower, there is a greater control of the heating process, the equipment dimensions are much smaller compared to the conventional heating techniques.

Previous experiments carried out by authors aimed at different types of microwave heating: direct, indirect and mixed (partially, direct-indirect). The indirect heating or the partially direct-indirect heating were obtained by placing between the material and the microwave generating source a screen made of silicon carbide (a material with high microwave susceptibility) in the form of a ceramic tube or crucible with a wall thickness varying between 3-20 mm. The large thickness (15-20 mm) of the screen wall completely absorbs the microwaves leading to indirect heating of the material by thermal radiation. The low thickness of the wall (3-5 mm) allows both its partial penetration by the microwave, and the partial absorption of the microwaves in the screen mass, achieving a mixed microwave heating. It has been experimentally found that the direct heating is excessively intense for the foaming process of the soda-lime glass (commercial container glass) used as a raw material, leading to much high heating rates of over 40 ºC/min, and severely destroying the internal structure of the material (Paunescu et al., 2017). Predominantly direct microwave heating (screen wall thickness: 3.5-5 mm) allowed optimal heating rates (15-25 ºC/min) for obtaining homogeneous porous microstructure of the cellular glass (Axinte et al., 2019). Research to determine the correlation between the wall thickness of the silicon carbide screen, and the proportion of its penetration by the microwave field has not been performed.

The experiments were performed in the company Daily Sourcing & Research on a 0.8 kW-microwave oven. The oven is of the type commonly used in the household for food preparation, having a single microwave power supply, the waveguide being provided in one of the side walls of the oven (Figure 1b). The main constructive modifications aimed at adaptations to ensure the operating conditions at high temperature (over 1000 ºC), and creating the possibility

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of measuring the process temperature by mounting a radiation pyrometer above the oven at about 400 mm (Figure 1a), which visualizes the surface of the material subjected to heating through holes in the upper metal wall of the oven, the ceramic lid of the enclosure containing the material, and the ceramic fiber mattress that thermally protects the lid. The powder mixture of raw material (glass waste, coal ash and silicon carbide as foaming agent), pressed into a mold and then released, is freely deposited on a 1 mm metal plate placed on a bed of ceramic fiber mattresses at the base of the oven. The material is covered with a 5 mm thick silicon carbide ceramic tube, and a ceramic lid of the same material provided with a hole with a diameter of 30 mm for measuring the temperature with the pyrometer. Both the wall of the ceramic tube and the ceramic lid are thermally protected on the outside with ceramic fiber mattresses. Given that the propagation of heat in the material mass takes place from the inside to the outside, as opposed to conventional heating processes, the thermal protection of the material is very important.

a *b*

Figure 1. Microwave equipment for manufacturing the cellular glass
a – overall image of the equipment; b – 0.8 kW-microwave oven.

As mentioned above, the raw material was composed of container glass waste (including green, amber and colorless glass), broken, ground in a ball mill, and sieved at a grain size below 180 µm as well as coal ash purchased from a Romanian thermal power station. The coal ash contained both fly ash captured in electrofilters and bottom ash. The grain size of the coal ash was below 200 µm. The chemical composition of the raw material is shown in Table 1.

Table 1. Chemical composition of raw material

Chemical composition	**Container glass waste, wt.%**			**Coal ash, wt.%**
	Colorless	Green	Amber	
SiO_2	71.7	71.8	71.1	46.5
Al_2O_3	1.9	1.9	2.0	23.7
CaO	12.0	11.8	12.1	7.9
Fe_2O_3	-	-	0.2	8.6
MgO	1.0	1.2	1.1	-
Na_2O	13.3	13.1	13.3	6.0
K_2O	-	0.1	0.1	4.1
Cr_2O_3	0.05	0.09	-	-
SO_3	-	-	0.05	-
Other oxides	0.05	0.01	0.05	-

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The chemical composition and the particle size of the coal ash make it suitable for the direct incorporation into a ceramic powder without prior processing. The ash partially replaces kaolin, feldspar and quartz. Although the iron oxide in the ash composition is a contaminant for the final product, negatively influencing its coefficient of thermal expansion (Yao et al., 2015; Kolberg & Roemer, 2001), during the microwave heating it has the role of absorbing electromagnetic radiation more efficiently at room temperature.

The silicon carbide with a fine granulation (below 63 µm) was selected as a foaming agent, being also a high microwave susceptible material. The silicon carbide (SiC) used in experiments is considered a very effective foaming agent capable of producing a foamed material with a very homogeneous cellular structure with controllable dimensions. Generally, the temperature range in which SiC is active is 950-1150 ºC. It reacts with oxygen in the oxidizing atmosphere of the oven, releasing CO_2 and CO according to the reactions below. The residual silicon oxide resulting from the reactions is incorporated into the mass of the material (Scarinci et al., 2005).

$SiC + 2O_2 = SiO_2 + CO_2$

$SiC + O_2 = SiO + CO$

Based on the results of several previous silicon carbide foaming experiments of glass waste associated with coal ash (Dragoescu et al., 2018), the following weight proportion ranges of these components were adopted: 9-11 wt.% coal ash, 2.8-3.6 wt.% silicon carbide and 85.5-88.2 wt.% container glass waste. Supplementary, after the homogeneous mixing of the powder mixture, about 14.5 wt.% water was added to facilitate the cold pressing of the mixture. Four experimental variants were adopted being shown in Table 2.

Table 2. Experimental variants for producing cellular glass

Variant	Container glass waste wt.%	Coal ash wt.%	Silicon carbide wt.%	Water addition wt.%
1	88.2	9.0	2.8	14.5
2	87.5	9.5	3.0	14.5
3	86.7	10.0	3.3	14.5
4	85.6	11.0	3.6	14.5

Results and Discussion

The thermal process of manufacturing the cellular glass took place in the microwave oven described above. Unlike the microwave oven used in the household, the mechanism of rotation of the heated material placed at the base of the oven was not functional, the sample having a fixed position relative to the microwave field, without affecting the homogeneity of heating and uniformity of pore distribution in the expanded material mass. The main functional parameters of the foaming process are presented in Table 3.

Table 3. Functional parameters of the foaming process.

Parameter	Variant 1	Variant 2	Variant 3	Variant 4
Dry/wet raw material amount, g	600/ 687	600/ 687	600/687	600/ 687
Foaming temperature, ºC	958	960	963	966
Heating duration, min	42	42.5	43	44.5
Average rate, ºC/min -heating -cooling	 22.4 5.9	 22.2 5.7	 22.0 5.8	 21.3 5.7
Cellular glass amount, g	582	583	582	580
Index of volume growth	2.4	2.6	2.9	3.4

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Specific consumption of electricity, kWh/kg	0.84	0.85	0.86	0.89

The cellular glass samples experimentally manufactured by microwave irradiation were analyzed in the laboratory to determine the physical, mechanical and microstructural characteristics using classical methods of analysis. The apparent density was measured by the gravimetric method (Manual, 1999) and the porosity was determined by the method of comparing the density of the compact material (after melting and then cooling) and the density of the porous material (Anovitz & Cole, 2005). 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 compressive strength was determined using an own small-scale device 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 hydrolytic stability of the porous material was determined using the standard procedure ISO 719: 1985. The water absorption was measured by the common method of the sample immersion in water. The porous microstructure of the samples was identified with a Smartphone Digital Microscope. To investigate the crystallographic structure of the samples, X-ray diffraction (XRD) was used according to the standard EN 13925-2: 2003 using a diffractometer Bruker-AXS D8 Advance with CuK α radiation. The main physical and mechanical characteristics of the cellular glass samples are shown in Table 4.

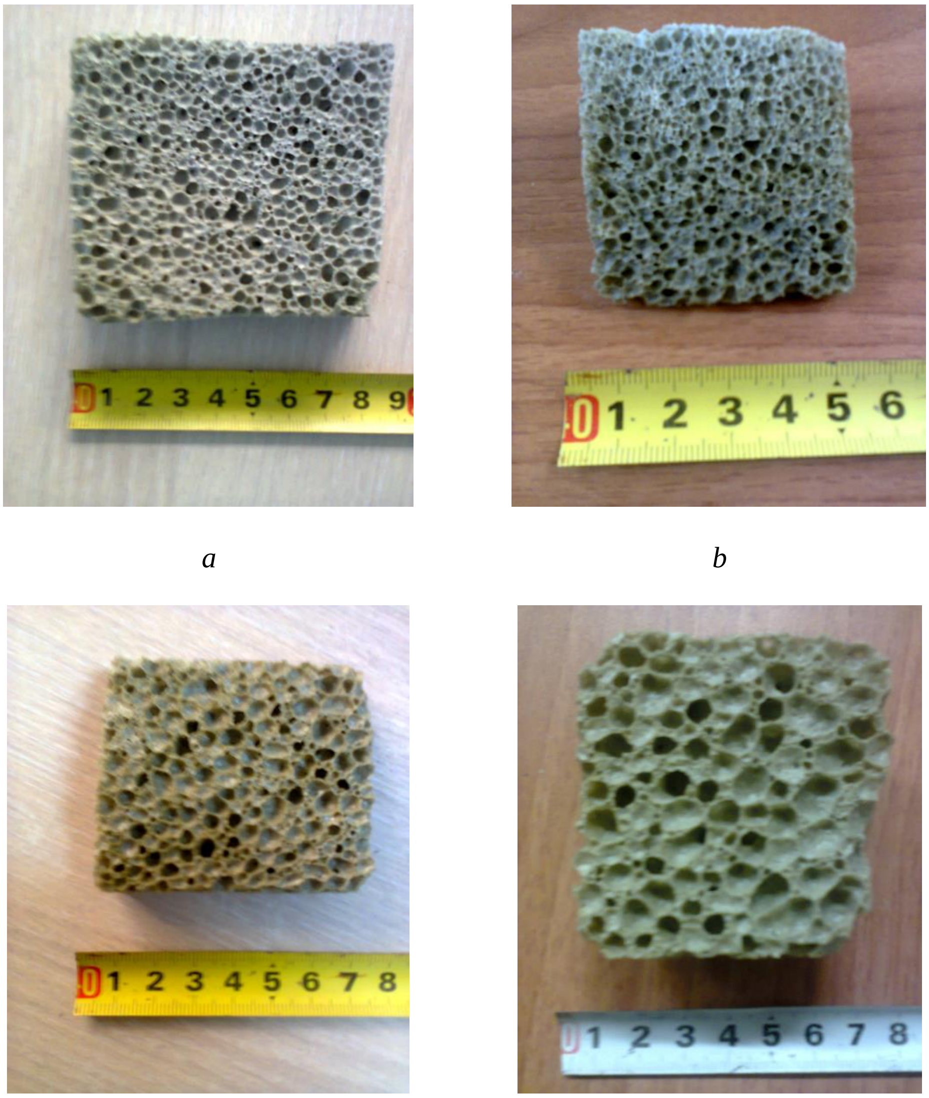
Table 4. Physical and mechanical characteristics of the cellular glass samples

Variant	**Apparent density g/cm^3**	**Porosity %**	**Thermal conductivity W/m·K**	**Compressive strength MPa**	**Water absorption %**	**Pore size mm**
1	0.32	85.5	0.060	1.34	0.4	0.4-1.0
2	0.29	86.8	0.055	1.30	0.6	0.7-1.3
3	0.25	88.6	0.049	1.27	1.0	0.8-2.1
4	0.22	90.0	0.043	1.23	1.2	1.7-2.5

According to the data in Table 3, the silicon carbide foaming process of the glass waste took place at the usual temperatures of conventional processes in the range of 958-966 C, the process duration being between 42-44.5 min. Due to the adoption of the predominantly direct microwave heating technique, the heating rate had optimal values (21.3-22.4 ºC/min), allowing to obtain homogeneous microstructures of the samples towards the upper limit recommended in conventional processes. The increase in volume by expanding the raw material was appropriate, being at least 2.4 times in the case of sample 1, and reaching 3.4 times in the case of sample 4. The measurements on the energy consumption showed the remarkable efficiency of the foaming process in the microwave field, the values of the specific energy consumption being between 0.84-0.89 kWh/kg. These values are comparable to those industrially made in high capacity conventional ovens. According to the considerations presented in the literature (Kharissova et al., 2010), an industrial-scale microwave equipment has a higher energy efficiency by up to 25% compared to a very low power microwave oven (0.8 kW) commonly used in household, of the type used in experiments described above. Therefore, it can be anticipated that an application on an industrial scale of experiments performed on a small-scale would allow the reduction of the specific energy consumption of the process up to 0.63-0.67 kWh/kg. The analysis of the data in Table 4 allowed the conclusion that the products manufactured by the unconventional microwave technique are, generally, similar to those industrially produced by conventional techniques. Thus, the main physical and mechanical characteristics of experimentally made cellular glass have the required properties to be used as thermal insulation material: low apparent density (0.22-0.32 g/cm^3), high porosity (85.5-

90.0%), low thermal conductivity (0.043-0.060 W/m·K), and acceptable compressive strength (1.23-1.34 MPa). Practically, the material is not water absorbent, the water absorption values being negligible.

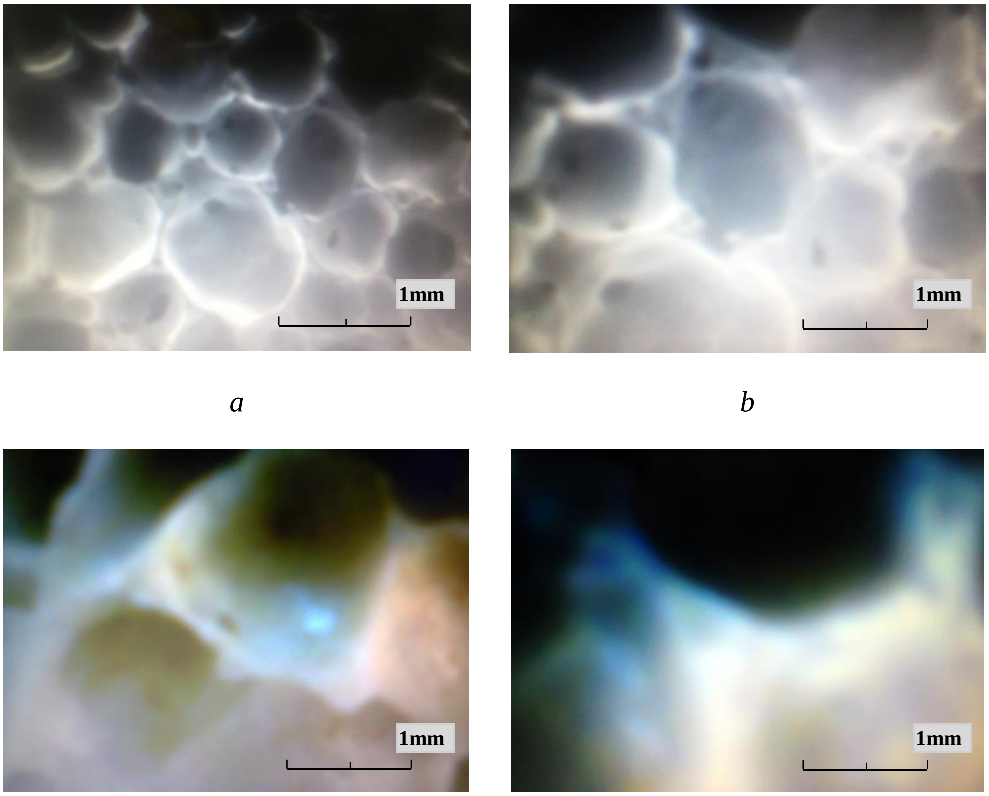
Pictures of section of the four cellular glass samples are shown in Figure 2. The uniformity of the pore distribution in the samples section is obvious. The pore size was identified by visualizing the microstructure of the samples (Figure 3). The smallest pore sizes belong to samples 1 (between 0.4-1.0 mm) and 2 (between 0.7-1.3 mm), and the largest correspond to samples 3 (between 0.8-2.1 mm) and 4 (between 1.7-2.5).



a *b*

c *d*

Figure 2. Pictures of the cellular glass samples
a – sample 1 heated at 958 ºC; b – sample 2 heated at 960 ºC;
c – sample 3 heated at 963 ºC; d – sample 4 heated at 966 ºC.



a *b*

c *d*

Figure 3. Microstructural images of the cellular glass samples
a – sample 1; b – sample 2; c – sample 3; d – sample 4.

The hydrolytic stability determination test for the four cellular glass samples, performed with a 0.01M HCl solution (0.15 ml) to neutralize the extracted Na_2O, indicated that the stability falls into the hydrolytic class no. 2 (the extracted Na_2O equivalent was between 37-50 µg. The XRD analysis to identify the crystalline phases of the cellular glass microstructure was performed on all the samples. The main crystalline phase identified after the heat treatment at 958-966 ºC was wollastonite-2M ($CaSiO_3$) and traces of silicon carbide (SiC).

Conclusion

The objective of the research that was the basis of the paper was the experimental production of cellular glass from glass waste, and coal ash as raw material, and silicon carbide as a foaming agent, using the unconventional microwave heating technique. The experiments were performed in the Daily Sourcing & Research company on a 0.8 kW-microwave oven of the type used in the household for food preparation, adapted for operation at high temperature (up to 1000 ºC). Although the microwaves have been known since the middle of the last century, their worldwide use has been limited to low-temperature drying and heating processes. The experimental research of high temperature heating processes using the microwave irradiation as a heat source is the originality of this paper. The microwave heating is completely different compared to the conventional heating. It is initiated in the core of the material, the heat propagating volumetrically to the peripheral areas, and it is also selective, acting only on the material subjected to heating. For these reasons, the microwave heating is fast and economical. The experiments presented in the paper aimed at the manufacture of porous materials with thermal insulating properties for the building construction similar in terms of quality to those industrially manufactured by conventional techniques, but with lower energy consumption. The results demonstrated the qualitative similarity of the products obtained by the unconventional technique compared to those conventionally manufactured. The specific energy consumption was low (0.84-0.89 kWh/kg) being comparable to the industrial one, but according to the literature, given the use of a microwave equipment on an industrial scale, this consumption could decrease by up to 25%.

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