ENHANCED MANUFACTURING TECHNIQUE OF CELLULAR GLASS-CERAMIC USING THE MICROWAVE ENERGY

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ABSTRACT: The paper presents experimental results aiming the enhanced manufacturing technique of cellular glass-ceramic using a mixed microwave heating method (predominantly direct and partially indirect). A SiC and Si_3N_4 ceramic tube with a wall thickness of 3 mm was used as a screen placed between the microwave radiation source and the material subjected to heating. The optimal sample was made by sintering at 980 ºC of 87.5% glass waste, 9.5% coal ash, 3% silicon carbide and 10% water addition. The product had the apparent density of 0.24 g/cm^3, porosity of 88%, thermal conductivity of 0.052 W/m·K and compressive strength of 1.25 MPa. The material microstructure was homogeneous, with pores uniformly distributed, having the size between 0.4-1.2 mm.
KEYWORDS: cellular glass-ceramic, microwave, foaming, glass waste, silicon carbide, mixed heating.

1. INTRODUCTION

Cellular glass-ceramic or glass-ceramic foam is a porous product obtained by a controlled crystallization of glass in a heat treatment at high temperature (800-1150 ºC) of various silicate waste (metallurgical slag, coal ash, incinerator ash, zinc hydrometallurgy waste , red mud, oil shale by-products, etc. [1-6]. By heat treatment, these wastes separately used or combined with each other, together with the glass waste and a common foaming agent (black carbon, calcium carbonate, silicon carbide, etc.) form a predominantly crystallized porous microstructure (between 50-90%), the rest being amorphous residual glass. Generally, the cellular glass-ceramic is not commonly manufactured industrially, being preferred glass foam, which uses as basic raw material only glass waste. Excepting the partially crystallized microstructure, there are no essential differences between the physical and mechanical properties of glass foam and cellular glass-ceramic. The material has low density, low thermal conductivity, sufficiently high compressive strength, it is fireproof, waterproof, non-deformable, chemically inert, non-toxic, resistant to the aggression of rodents, insects, bacteria, acids, etc. [1, 2, 4]. The field of application of the glass-ceramic foams and the glass foams is similar including thermal and sound insulators, lightweight structural components, filters, absorbers, floor, wall, roof and ceiling insulations, architectural panels, road and railway constructions, foundations, drainages, sports grounds, etc. [2].

Worldwide, there are numerous experimental tests for the small-scale manufacture of glass-ceramic foams, their results being presented in the literature.

In recent years, the authors' concern has been focused on the technique of manufacturing cellular glass-ceramic using glass waste and coal fly ash as raw materials and silicon carbide as a highly effective foaming agent. The arguments of this decision are the availability of the two wastes used as raw material and the foaming ability of silicon carbide, especially since the composition of glass and coal ash as well as silicon carbide favors the absorption of microwave radiation.

A light cellular glass ceramic was produced [4] from a mixture of silicate waste, composed of 20% coal ash and 80% post-consumer container glass waste. The method of sintering the powder mixture was used, in which 2% silicon carbide was added as a foaming agent. The apparent density of the expanded material was in the range of 0.2 - 0.4 g/cm^3 and the porosity had values between 70 - 90%. The optimal values of the sintering temperature of the material (between 1000 - 1050 ºC) were experimentally determined. In this temperature range the porosity value was of about 75% and the best uniformity of pore distribution was obtained. The porous products had the compressive strengths around 1.5 MPa and showed a relatively high resistance to thermal shock. The main crystalline phase identified by XRD analysis was

wollastonite and traces of silicon carbide. Cristobalite was not detected in these samples, despite the high proportion of silica in the raw material mixture.

A similar experiment [7], using the same types of raw materials (glass waste and coal ash) and silicon carbide as a foaming agent in a lower weight ratio of only 1%, sintered at 950 ºC at a average heating rate of 23.2 ºC/min, led to the obtaining of a cellular glass-ceramic with an apparent density of 0.18 - 0.35 g/cm^3 and a compressive strength of 0.9 - 1.8 MPa. The distribution of the pores was homogeneous, with dimensions between 1-3 mm.

Sintering at 950 ºC for 20 min of a powder mixture consisting of glass waste, coal fly ash (50-70%) and silicon carbide as a foaming agent led to the production of a light cellular glass-ceramic with a porosity of 81.55%, a bulk density of 0.267 g/cm^3 and a compressive strength of 0.98 MPa. The growth rate of the expanded sample was very high, reaching 5.81. The crystalline phases of the samples were identified by XRD analysis, being mulite and cristobalite [8].

The experiments described above were performed by conventional heating (electrical resistances or fossil fuel consumption). The specific energy consumption has not been specified in the literature, but theoretically it was very high due to the high thermal requirement, the small quantities of raw material and the low energy efficiency of the experimental equipment.

Since 2016, the Romanian company Daily Sourcing & Research Bucharest has started to test different cellular glass types using low power-experimental microwave equipment. According to the paper [9], cellular glass-ceramics were made from container glass waste, coal fly ash and silicon carbide as a foaming agent. The tests were performed on a 5 kW-microwave reactor having a silicon carbide crucible with a wall thickness of 20 mm placed inside the reactor cavity. The material powder mixture was pressed into a metal mold deposited on the bottom of the ceramic crucible. The supply of electromagnetic waves was performed through magnetron waveguides mounted on the side wall of the reactor and respectively, at its base. The weight ratio of glass waste varied between 77-95%, that of fly ash between 0-18.5% and the silicon carbide proportion was 4.5-5.0%. The total amount of materials loaded into the metal mold was between 59-160 g and the sintering/foaming temperature had values between 957-995 ºC. The value of the specific energy consumption was very high (between 45.2-180.4 kWh/kg) due to an excessively high heat loss to the outside of over 70%. The main physical and mechanical characteristics of the samples were: apparent density between 0.35-0.55 g/cm^3, porosity between 61.6-81.6% and compressive strength between 1.3-1.5 MPa.

Other experiments aimed at manufacturing cellular glass-ceramics [9] were performed on a 0.8-kW microwave oven commonly used in the household at the food preparation. Similar proportions of raw material and foaming agent were loaded and pressed into a ceramic mold made of SiC and Si_3N_4 (80/20 ratio) with a wall thickness of 5 mm covered with a lid from the same material. A much more efficient thermal protection of the powder material was achieved with several layers of ceramic fiber mattresses. The sintering/foaming temperature varied between 961-990 ºC and the specific energy consumption was much reduced, being in the range 5.0-6.6 kWh/kg. The main physical and mechanical features of the cellular glass-ceramic samples were: apparent density between 0.32-0.34 g/cm^3, porosity between 82.0-83.2% and compressive strength between 1.2-1.4 MPa.

The same research team from Romania improved the manufacturing technique of cellular glass-ceramics in a 0.8 kW-microwave oven [10] using 85.0-89.0 wt.% glass waste, 9.0-10.5 wt.% coal fly ash, 2.0-4.5 wt.% silicon carbide and 9.0 wt.% water addition. The powder mixture moistened with water was pressed into a metal mold and then extracted from the mold and placed freely on a metal plate placed above the bed of ceramic fiber mattresses at the base of the oven. In order to optimize the microwave heating of the material, the predominantly direct and partially indirect mixed heating technique was adopted by placing a ceramic tube made of SiC and Si_3N_4 (80/20 ratio) provided with a lid made of the same material. The wall thickness of the ceramic tube was 3.5 mm considered optimal so the penetration of the wall by electromagnetic waves to be achieved with a moderate intensity so that the internal structure of the material be not affected by the direct contact with microwaves. Also, the microwave fields is partially absorbed into the ceramic tube mass, this being rapidly and efficiently heated and constituting a second heat transfer source to the material by thermal radiation. According to the literature [10], the sintering/foaming temperature was between 972-984 ºC and the samples had the apparent density between 0.17-0.27 g/cm^3, porosity between 87.7-92.3%, and compressive strength between 1.15-1.30 MPa. The specific energy consumption was in the range 1.46-1.84 kWh/kg. Generally, the uniformity of pore distribution was good, the pore size being between 1-4 mm, the lower

values corresponding to the silicon carbide ratio below 3.5 wt.%.

The current paper constitutes a continuation of research aiming at producing cellular glass-ceramics by the nonconventional microwave heating technique having the objective of reducing the specific energy consumption compared to the latest experimental results in conditions of obtaining products with physical, mechanical and microstructural characteristics according to the requirements for their use as thermal insulating materials.

2. METHODS AND MATERIALS

2.1 Methods

In principle, the method adopted for the production of cellular glass-ceramics aiming at reducing the specific energy consumption and keeping the physical, chemical and microstructural characteristics is similar to that used by the authors in the paper [10]. The method is based on the application of mixed microwave heating by placing a ceramic screen of a material with high microwave susceptibility capable to allow the passage of the electromagnetic wave radiation in a proportion that does not cause destruction of the internal structure of the heated material and at the same time, to absorb radiation by heating quickly and intensely. In this way, mainly a direct microwave heating acts on the material being initiated in its core, which propagates from the inside to the peripheral areas and also an indirect heating acts by the thermal radiation of the hot inner surface of the screen to the material surface. Own previous experiments have determined an optimal thickness of the ceramic screen of 3.5 mm. In the current paper, this thickness value of the screen was reduced to 3.0 mm. The screen is a SiC and Si_3N_4 ceramic tube with an outer diameter of 125 mm and a height of 100 mm placed on the bed of ceramic fiber mattresses at the base of the oven protecting the material subjected to heating against the high intensity of the microwave field. The thickness and quality of the insulating layers of ceramic fiber that wrap the outer wall of the ceramic tube and the lid of the tube have a very important role for the energy efficiency of the process. Thus, both the directly microwave heated material and the hot outer surface of the ceramic tube are effectively protected with ceramic fiber, greatly reducing the heat loss to the environment and implicitly, saving primary energy. In Figure 1 is presented the experimental microwave equipment being shown the 0.8 kW-microwave oven (a) and the ceramic fiber thermal protection of the ceramic tube including the heated material (b).

a b

Figure 1. Experimental microwave equipment
a – 0.8 kW-microwave oven; b – ceramic fiber thermal protection.

According to the literature [2, 4], the basic principle of foaming the glass based-raw material with silicon carbide as a foaming agent is the release of carbon dioxide (or carbon monoxide) in the viscous mass of the thermally softened material resulting by oxidation reactions of the foaming agent in the oxidizing atmosphere of the oven. The oxidation reaction (1) and (2) occur in the temperature range 950-1150 ºC.

$$SiC + 3/2O_2 = SiO_2 + CO \quad (1)$$

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

There should be a good correlation between the softening temperature of the powder mixture and the temperature at which the above chemical reactions occur so that the gases released in the form of

bubbles to remain trapped in the viscous material and then, by cooling, to form a specific porous structure.

2.2 Materials

The raw material used in experiments were container glass waste (between 87-92 wt.%) and coal fly ash (between 3-11 wt.%). Also, silicon carbide (between 2-5 wt.%) as a foaming agent was used. The glass waste including colorless, green and amber glass in equal weight proportions was broken, ground in a ball mill and sieved below 80 μm. The average chemical composition of the glass mixture included: SiO_2 (73.19%), Al_2O_3 (1.44%), Na_2O (12.85%), K_2O (0.75%), CaO (10.20%), MgO (1.34%), Fe_2O_3 (0.06%), SO_3 (0.19%), TiO_2 (0.04%) and BaO (0.03%) [11].

The coal fly ash used in experiments was provided by Paroseni thermal power station having the following chemical composition, according to the supplier's data: SiO_2 (46.5%), Al_2O_3 (23.7%), CaO (7.9%), MgO (3.2%), Na_2O (6.0%), K_2O (4.1%) and Fe_2O_3 (8.6%). The grain size of coal ash was below 63 μm after grinding in a ball mill.

The silicon carbide purchased from the market had the grain size below 5 μm.

3. RESULTS AND DISCUSSION

3.1 Results

The experimental glass-ceramic cellular manufacturing process was performed in the company Daily Sourcing & Research on the 0.8 kW-microwave oven described above. Four experimental variants were adopted (Table 1) including container glass waste, coal fly ash, silicon carbide and a constant weight ratio of water addition to facilitate the powder material cold pressing.

Table 1. Experimental variants for producing cellular glass-ceramics

Variant	Glass waste wt.%	Coal fly ash wt.%	Silicon carbide wt.%	Water addition wt.%
1	87.0	11.0	2.0	10.0
2	87.5	9.5	3.0	10.0
3	87.4	8.8	3.8	10.0
4	92.0	3.0	5.0	10.0

The main functional parameters of the sintering/foaming process are shown in Table 2.

Table 3 presents the main physical, mechanical and microstructural characteristics of the cellular glass-ceramic samples. The determination of the samples characteristics was performed using common methods of analysis. The apparent density was measured by the gravimetric method [12] and the porosity was calculated by the method of comparing the true and apparent density [13]. The compressive strength was determined using a Stable Micro Systems TA XT Plus Texture Analyzer and the thermal conductivity by the guarded-comparative-longitudinal heat flow (ASTM E1225-04 standard). The water absorption was measured by the water immersion method (ASTM D570 standard). The samples microstructure was examined with a Smartphone Digital Microscope.

Table 2. Functional parameters of the sintering/foaming process

Variant	Dry/wet raw material amount g	Sintering/ foaming temperature °C	Heating time min	Average rate, °C/min		Index of volume growth	Cellular glass-ceramic amount g	Specific energy consumption kWh/kg
				Heating	Cooling			
1	500/550	985	45	21.4	5.3	2.60	482	1.09
2	500/550	980	42	22.9	5.6	2.80	480	1.02
3	500/550	974	39	24.5	5.4	2.95	482	0.94
4	500/550	965	36	26.3	5.3	3.40	481	0.87

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

Variant	Apparent density g/cm^3	Porosity %	Compressive strength MPa	Thermal conductivity W/m·K	Water absorption %	Pore size mm
1	0.26	87.0	1.30	0.059	1.4	0.1 – 0.5
2	0.24	88.0	1.25	0.052	0.9	0.4 – 1.2
3	0.23	88.5	1.23	0.049	0.9	0.9 – 1.5
4	0.21	89.4	1.10	0.046	1.5	2.8 – 3.4

Examining the data in Table 2, the high values of the heating rate, but in controlled limits between 21.4-26.3 ºC/min as well as the low level of the specific energy consumption (0.87-1.09 kWh/kg) are remarkable. The use of the ceramic tube with a wall thickness of 3 mm as a screen for moderating the intensity of the direct contact of microwaves with the material subjected to heating allowed to achieve optimal heating rates for the cellular glass-ceramics manufacturing process, also recommended for conventional processes [1, 4]. The specific energy consumption was very low, the manufacturing process described in the current paper being clearly more efficient compared to the similar one presented in the paper [10].

The characteristics of the glass-ceramic cellular samples obtained by the nonconventional technique of microwave heating shown in Table 3, confirm that this technique allows the manufacture of some products similar to those made by conventional techniques. Apparent density between 0.21-0.26 g/cm^3, porosity between 87.0-89.4%, thermal conductivity in the range 0.046-0.059 W/m·K, compressive strength between 1.10-1.30 MPa, uniformity of the pore distribution and their size in very low limits for samples 1-3 (made with 8.8-11.0 wt.% coal ash and 2.0-3.8 wt.% silicon carbide) in the range 0.1-1.5 mm, are optimal for using this material type as thermal insulating in construction.

Pictures of section cellular glass-ceramic samples made in microwave field are shown in Figure 2 and microstructural images of the samples are presented in Figure 3.

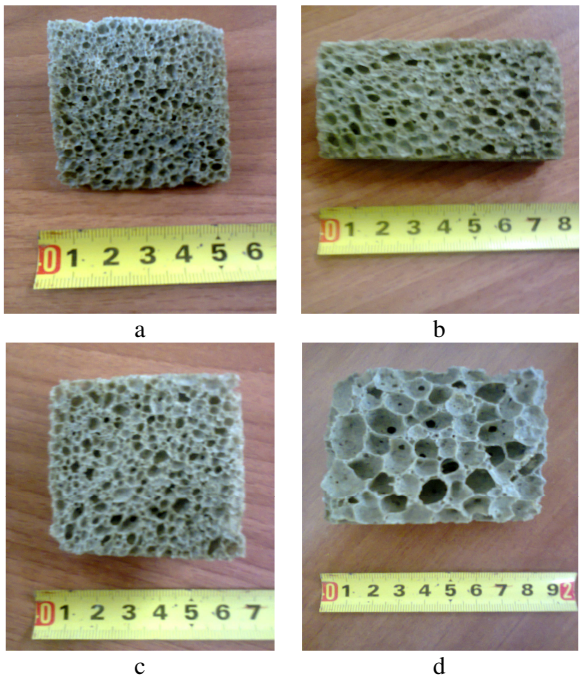

Figure 2. Pictures of section cellular glass-ceramic samples
a – sample 1, heated at 985 ºC; b - sample 2, heated at 980 ºC;
c – sample 3, heated at 974 ºC; d - sample 4, heated at 965 ºC.

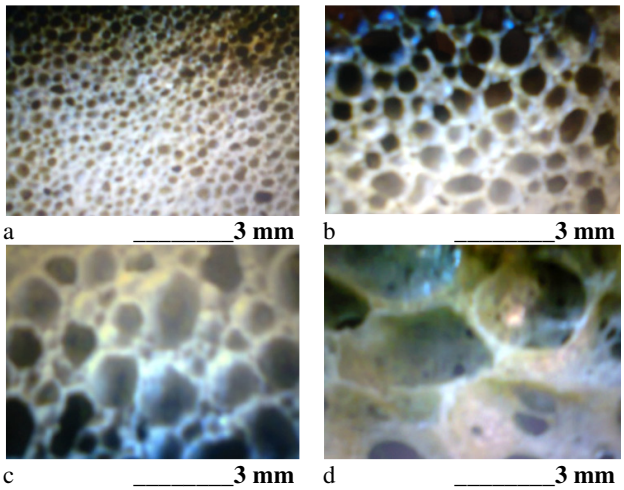

Figure 3. Microstructural images of the cellular glass-ceramic samples
a – sample 1; b – sample 2; c – sample 3; d – sample 4.

Analyzing the pictures in Figure 2, it is observed that the four cellular glass-ceramic samples have homogeneous microstructures with uniform pore distribution in section. As the silicon carbide ratio increases and the coal ash ratio decreases, the pore size is constantly increasing, from 0.1-0.5 mm (sample 1) to 0.4-1.2 mm (sample 2), 0.9-1.5 mm (sample 3). and 2.8-3.4 mm (test 4). It is clear that by increasing the silicon carbide ratio up to 5 wt.%, the sample microstructure is characterized by large pores, which affects the compressive strength of the material. Simultaneous reduction of the coal ash ratio up to 3 wt.% leads to a decrease in the sintering/foaming temperature. Examining the images of the samples microstructure in Figure 3, it is observed that in the case of sample 4 there is a clear tendency of communication between the neighboring cells and the formation of a semi-open microstructure. The other samples have small closed cell microstructures.

3.2 Discussion

The mixed microwave heating (predominantly direct and partially indirect) using a ceramic tube with high microwave susceptibility, with a wall thickness of 3 mm, has experimentally proven its superior energy efficiency compared to other types of microwave heating and conventional heating techniques. The specific energy consumption of the experimental process was between 0.87-1.09 kWh/kg. According to the literature [14], the use of an industrial scale microwave equipment could have an energy efficiency up to 25% higher than the 0.8 kW-microwave oven on which the tests were performed.

In terms of quality, the cellular glass-ceramic samples had physical, mechanical and microstructural characteristics similar to those of products obtained by conventional techniques being suitable for use as thermal insulating materials in construction. Of the four experimental samples, the optimal sample was considered sample 2 made of 87.5% glass waste, 9.5% coal ash, 3% silicon carbide and 10% water addition. The product had the apparent density of 0.24 g/cm^3, thermal conductivity of 0.052 W/m·K and compressive strength of 1.25 MPa, the pore size being between 0.4-1.2 mm.

4. CONCLUSION

The research objective was improving the energy efficiency of the manufacturing in microwave field of cellular glass-ceramics. The solution was the use of a SiC and Si_3N_4 ceramic tube with the wall thickness of 3 mm, that allows a mixed heating (predominantly direct and partially indirect) of the raw material powder mixture. The optimal sample was made by sintering at 980 ºC of 87.5% glass waste, 9.5% coal ash, 3% silicon carbide and 10% water addition. The cellular glass-ceramic had the apparent density of 0.24 g/cm^3, thermal conductivity of 0.052 W/m·K and compressive strength of 1.25 MPa, the pore size being between 0.4-1.2 mm.

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