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Glass Foam Made with Silicon Nitride and Manganese Oxide by Microwave Irradiation

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

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

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

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Abstract

A high mechanical strength (6.1 MPa) glass foam was produced by sintering/foaming at 830 ºC in an experimental 0.8 kW-microwave oven. The basic raw material was a colorless flat glass waste and the foaming agent was Si3N4 powder (2 wt.%). As an oxygen supplying agent, a MnO2 powder (3.1 wt.%) was used. The main physical, mechanical, thermal and morphological characteristics of the optimal sample were: apparent density of 0.47 g/cm3, porosity of 77.6%, thermal conductivity of 0.105 W/m·K, compressive strength of 6.1 MPa and pore size between 0.15-0.40 mm. The optimal glass foam sample has the required characteristics of a thermal insulation material usable under mechanical stress conditions in civil engineering. The originality of the paper is the application of the unconventional microwave heating technique, faster and more economical, unlike the other papers in the same area published in the literature, followers of the traditional conventional heating technique.

Introduction

The beginning of the global energy (hydrocarbons) crisis from the 1970' years as well as the atmosphere pollution with greenhouse gases (mainly carbon dioxide) and implicitly, the tendency of the terrestrial climate overheating led to the adoption of a new attitude towards energy and environmental protection. Thus, since the last decades of the twentieth century, recycling the various types of waste (plastic, metal, paper, glass, etc.) with direct energy and environmental involvement has become a major concern of the world and especially of developed countries and those with an intense development rate, as the main energy consumers and waste generators. High consumers of thermal energy with massive emissions of carbon dioxide into the atmosphere from the building materials industry, chemical industry and others (de Bruin et al., 2020) have already adopted measures for the partial replacement of their basic raw materials with recycled material waste.

Glass waste constitutes a large amount available worldwide and with a constantly growing annual generation rate. Several international companies with facilities in Europe and China (mainly Misapor and Pittsburgh Corning) (Dragoescu et al., 2018a) already produce on an industrial scale several types of glass foam from glass waste, with thermal and/or acoustic insulating properties, with mechanical strengths in a wide range of values (from moderate to high) covering wide fields of application in the construction sector as replacers of some traditional materials existing on the market. Also, worldwide there are concerns on an

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experimental scale for the diversification of manufacturing recipes with new waste types as raw materials and additives, improved techniques and advanced characteristics of foam products, the literature providing information in this area. The most common type of recycled glass waste used in glass foam manufacturing processes as a basic raw material is post-consumer container glass, which is a soda-lime glass. All common foaming agents (carbon black, petroleum coke, graphite, coal, calcium carbonate, silicon carbide, etc.) can be used in this process. According to the literature (Scarinci et al., 2005), carbon-containing foaming agents are most commonly used in industrial processes. The temperature at which the sintering and foaming of soda-lime glass occurs is in the range of 800-900 ºC, the foaming gases (CO2 and CO) resulting from the oxidation of carbon in the oxidizing atmosphere of the oven.

Flat glass waste is also used in industrial processes, but only in combination (about 10 wt.%) with the commercial container glass waste for foam glass gravel manufacture (Geocell, 2016). It was found that when used alone, the flat glass waste forms an inhomogeneous microstructure regardless of the foaming agent type, using conventional heat treatment techniques. However, experiments performed in a low power microwave oven in the Romanian company Daily Sourcing & Research allowed to obtain foamed products with a very good microstructural homogeneity using silicon carbide together with coal ash (Dragoescu et al., 2018b) or calcium carbonate (Dragoescu et al. 2018c). The other physical, thermal and mechanical characteristics were suitable for the use of glass foams as thermal insulation material in civil engineering.

Another glass waste type tested on an experimental scale as a raw material in the process of making glass foam is cathode ray tube (CRT) glass out of use. According to (Scarinci et al., 2005), a suitable foaming agent for foaming the CRT glass waste is calcium carbonate. The products are lightweight, with very fine porosity and microstructural homogeneity. Clam shell powder (between 2-10 wt.%) with high calcium carbonate content was also experimentally used to manufacture glass foams in the shape of pellets by sintering and foaming the CRT glass waste at 700-750 ºC (Lunip et al., 2016). Several papers presented in the literature (Petersen et al., 2015; Kőnig et al., 2016) refer to the use of manganese oxide (MnO2) as a foaming agent in experimental processes for the production of glass foam from CRT glass waste. A technique for producing glass foam in the form of pellets presented in (Petersen et al., 2014) consists in the sintering at 750 ºC of a mixture of CRT glass waste and sodium carbonate (Na2CO3) in a weight ratio of 14% as a foaming agent. A mixture of CRT glass waste, dolomite (CaMg (CO3)2) and CaCO3 as foaming agents, Na-bentonite and Ca-bentonite as pelletizing agents and a low addition of alumina (Al2O3) was pelletized and sintered at 800 ºC resulting porous products with very low density and high mechanical strength usable as building materials (Mucsi et al, 2013). Other types of foaming agents used for the experimental manufacture of glass foam from CRT glass waste were fly ash (Barbosa et al., 2015) as well as a mixture of carbon (C), titanium oxide (TiO2), MnO2 and aluminum nitride (AlN) (Laur et al., 2017).

Another foaming agent (silicon nitride Si3N4) considered very effective was used for foaming the flat glass waste together with an oxygen-supplying agent (MnO2), in addition to the oxygen in the oven atmosphere (Llaudis et al., 2009). As the MnO2/Si3N4 molar ratio increases, both the bulk density and the compressive strength decrease. Also, with the increase of the heating temperature, the density value of the glass foam slightly decreases, while the decrease of the compressive strength value is more accentuated. According to (Garcia Ten et al., 2011), the association of the foaming agent oxidation (SiC, Si3N4 or AlN) with the reduction of multiple valence-metal oxides (MnO2, Fe2O3 or CeO2) leads to the highest efficiency of the foaming process of a soda-lime glass waste and better microstructural homogeneity. It was experimentally found that the use of Si3N4 together with MnO2 is the best combination between the expansion degree and compressive strength of the glass foam. According to the paper (Du et al., 2019), mixed surfactants (Khan & Marques, 2019) were used to manufacture

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porous Si3N4 ceramic foams by the direct foaming method. Ceramic foams with very high porosity, pore size between 0.14-0.24 mm and compressive strength between 0.85-5.38 MPa were obtained by varying the mixture of surfactants between 0.1-0.4 wt.% and a solid content between 22-30 vol.%.

The aim of the current paper is the experimental manufacture of a glass foam from flat glass waste using Si3N4 as a foaming agent and MnO2 as an oxygen supplying agent. Unlike most glass foam manufacturing processes in the world that use conventional heating techniques, the experimental process described further is based on the unconventional microwave heating technique, constituting the originality of the work.

Methods

The basic principle of foaming the glass waste is the release of a gaseous compound by the foaming agent in the softened mass of the glass, which receives it in the form of gas bubbles. The suitable viscosity of molten glass trappes the bubbles. By cooling the bubbles turn into pores forming a typical porous structure (Scarinci et al., 2005).

The stages that make up the foaming process of the glass waste using Si_3N_4 as a foaming agent and MnO_2 as an oxygen supplying agent are the following:

$$4MnO_2 = 2Mn_2O_3 + O_2 \quad (1)$$

$$6Mn_2O_3 = 4Mn_3O_4 + O_2 \quad (2)$$

$$Si_3N_4 + 7O_2 = 3SiO_2 + 4NO_2 \quad (3)$$

The decomposition of MnO_2 (1) occurs at about 483 ºC with the maximum peak at about 590 ºC and the decomposition of Mn_2O_3 (2) occurs at about 650 ºC reaching the maximum peak at about 800 ºC (Terayama & Ikeda, 1983). The oxygen released by reactions (1) and (2) participates in the oxidation reaction of Si_3N_4. According to (Yurkov, 2015), the oxidation reaction of Si_3N_4 occurs in the temperature range 800-900 ºC. In case of foaming with Si_3N_4 the gaseous compound released in the molten glass mass is NO_2 (3).

The used experimental equipment was composed of the 0.8 kW-microwave oven (1) of the type commonly used in the household with a single microwave generator (8), but adapted for operation at high temperature (up to 1200 ºC), a ceramic tube (5) made of a material with high microwave susceptibility (a mixture 80/20 of SiC and Si_3N_4) to reduce the too much intensity of the contact of microwave field with the material subjected to heating, a ceramic lid (2) of the same material as the tube to close the open upper area and a metal plate (4) on which it is deposed freely the pressed material (3), placed on a bed of ceramic fiber mattresses. Thick layers of ceramic fiber mattresses (7) thermally insulate the heated material, the ceramic tube and the lid. The role of the ceramic tube is to allow the predominant (but not total) penetration of the microwave field for a direct heating of the material, but also to partially absorb the microwave flow for an indirect heating of the material. Thus, a mixed heating can be performed in order to protect the internal structure of the glass against the too intense attack of the microwave flow. The direct heating is initiated in the core of the microwave irradiated material by converting the electromagnetic waves power into heat. The heat is then volumetrically transmitted from the inside of the material to its outer areas (Jones et al., 2002). The selectivity of the direct heating (Kitchen et al., 2014) which allows the heating of only the irradiated material ensures a higher energy efficiency of the unconventional technique compared to conventional heating techniques. The indirect heating is of the conventional type through the thermal radiation of the ceramic tube wall. The thermal control of the process is performed with a radiation pyrometer (9) which visualizes the irradiated material through 30 mm-holes

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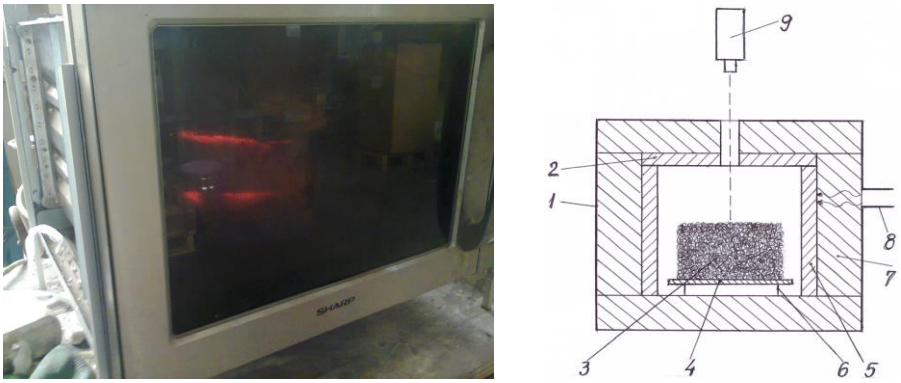
provided in the upper metal wall of the oven, ceramic lid and the ceramic fiber-thermal insulation material.



A **B**

Figure 1. Images of the experimental microwave equipment

A – 0.8 kW-microwave oven picture; B – constructive scheme of the equipment: 1 – microwave oven: 2 – ceramic lid; 3 – pressed mixture; 4 – metal plate; 5 – ceramic tube; 6 – metal support; 7 – ceramic protection; 8 – waveguide; 9 – pyrometer.

The materials used in the experimental process of making glass foam were: colorless flat glass waste from the window glass cullet recycling as a raw material, MnO_2 as an oxygen supplying agent and Si_3N_4 as a foaming agent. The flat glass waste was broken, ground in a ball mill and sieved at the grain size below 100 μm. The chemical composition of the window glass is shown [6] in Table 1. MnO_2 was purchased from the market at a grain size of 40-50 μm and was used in experiments without further mechanical processing. Si_3N_4 with a fine granulation (below 10 μm) was also purchased from the market, thus being used.

Table 1. Chemical Composition of Window Glass

SiO_2	CaO	MgO	Na_2O	Al_2O_3	Fe_2O_3
71.5	7.9	3.6	15.6	1.2	0.2

Considering the effectiveness of combining the oxidation reaction of the foaming agent (Si_3N_4) with the reduction reaction of a metal oxide with multiple valence (MnO_2 type) to obtain a good microstructural homogeneity of glass foam observed in some papers presented in the literature, four experimental variants were adopted by authors for their testing in the 0.8 kW-microwave oven. According to Table 2, the foaming agent ratio was kept constant at 2 wt.%, while the MnO_2 ratio varied between 2.0-5.5 wt.%. Flat glass waste was used in weight proportions between 92.5-96.0%. A constant amount of water addition representing 14.0 wt.% was used to facilitate the cold pressing of the powder mixture.

Table 2. Experimental Variants Composition

Variant	Colorless flat glass waste wt.%	MnO_2 wt.%	Si_3N_4 wt.%	Water addition wt.%
1	96.0	2.0	2.0	14.0
2	94.9	3.1	2.0	14.0
3	93.8	4.2	2.0	14.0
4	92.5	5.5	2.0	14.0

The experimental process of glass foam manufacturing took place in the company Daily Sourcing & Research on the 0.8 kW-microwave oven described above. The process functional

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parameters including the quantities of dry raw material and glass foam, sintering temperature and process duration, average heating and cooling rate, index of volume growth and specific energy consumption are shown in Table 3.

Table 3. Main Functional Parameters of the Foaming Process

Parameter	Variant			
	1	2	3	4
Dry raw material/glass foam amount (g)	480/465	480/467	480/470	480/468
Sintering/foaming temperature (ºC)	815	830	845	860
Heating time (min)	35	37	40	44
Average heating rate (ºC/min)	22.7	21.9	20.6	19.1
Average cooling rate (ºC/min)	6.8	6.4	6.7	6.6
Index of volume growth	1.35	1.50	1.70	1.80
Specific energy consumption (kWh/kg)	0.78	0.83	0.89	0.98

According to the data in Table 3, increasing the weight ratio of MnO_2, which favorably influences the glass foaming process, the value of the sintering/foaming temperature increased from 815 to 860 ºC. Implicitly, the process time increased also in the range 35-44 min. Due to the type of mixed microwave heating, the average heating rate was high (between 19.1-22.7 ºC/min) significantly exceeding the level commonly used in similar conventional processes (10-15 ºC/min) (Scarinci et al., 2005). The average cooling rate adopted by the authors was slightly higher (6.4-6.8 ºC/min) compared to the rates used in conventional processes (5.0-5.5 ºC/min). The specific energy consumption had very low values (between 0.78-0.98 kWh/kg) especially in the case of manufacturing glass foams with lower consumption of MnO_2 which determines a lower value of the sintering/foaming temperature and therefore shorter durations of the heating process.

The ensemble of the physical, thermal, mechanical and morphological characteristics of the analyzed products has a special importance when evaluating the optimal foamed product. The investigation of these characteristics of the four experimentally made glass foams was performed using common analysis techniques. The apparent density was measured by the gravimetric method (Manual, 1999) and the porosity was calculated by the method of comparing the true and apparent density (Anovitz & Cole, 2005). The compressive strength was determined using a TA.XTplus Texture Analyzer of Stable Micro Systems (ASTM C552-17) and the thermal conductivity was measured by the heat-flow meter method (ASTM E1225-04). The water absorption was determined by the water immersion (for 24 hours) method (ASTM D570). The products microstructure was examined with an ASONA 100X Zoom Smartphone Digital Microscope. The main physical, thermal, mechanical and morphological characteristics of glass foam samples are presented in Table 4.

Table 4. Main Physical, Thermal, Mechanical and Morphological Features of Glass Foam Samples

Variant	Apparent density g/cm^3	Porosity %	Thermal conductivity W/m·K	Compressive strength MPa	Water absorption %	Pore size mm
1	0.57	72.9	0.123	7.8	3.5	0.10-0.35
2	0.47	77.6	0.105	6.1	4.9	0.15-0.40
3	0.36	82.9	0.085	3.2	5.0	0.20-0.50
4	0.38	85.6	0.089	4.9	6.6	0.35-0.55

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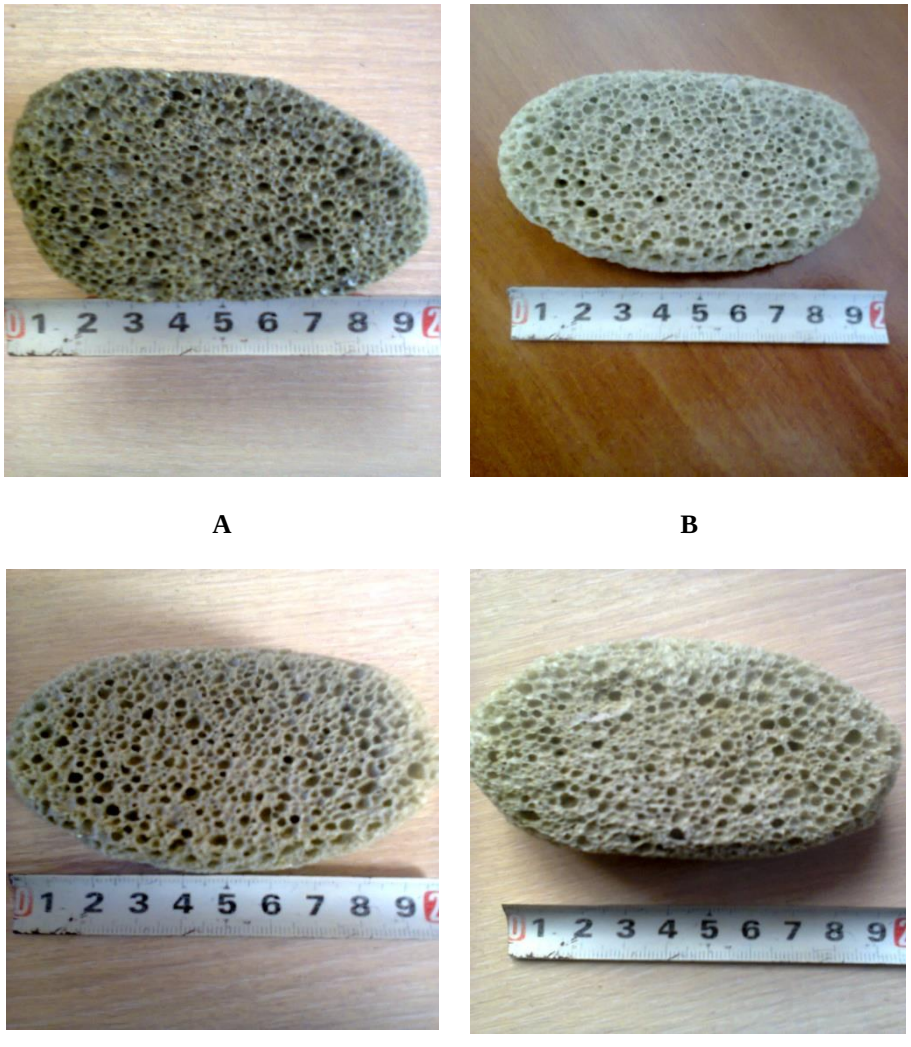
Analyzing the data in Table 4, it can be observed an improvement of the thermal insulating properties of the samples by decreasing the value of the apparent density and of the thermal conductivity due to the addition of MnO_2. However, in the case of the sample corresponding to variant 4 (with 5.5 wt.% MnO_2) this tendency is slightly modified, the values of the two physical and thermal characteristics increasing. This change can be explained by the cells coalescence and the formation of connection channels in their thicker walls, which are observed at higher values of material porosity (in this case at a porosity of 95.6%). In Figure 3D the existence of the interconnection channels in the cell walls can be clearly seen. As a consequence, the compressive strength, which showed a tendency to decrease its value from 7.8 MPa (variant 1) up to 3.2 MPa (variant 3), increases again to 4.9 MPa in the case of variant 4. The water absorption measured in the porous material was relatively low (between 3.5-6.6%). Pictures of the cross section and the microstructural configuration of the four glass foam samples are shown in Figure 2 and Figure 3, respectively.



A **B**

C **D**

Figure 2. Cross section of glass foam samples
A – sample 1 sintered at 815 ºC; B – sample 2 sintered at 830 ºC;
C – sample 3 sintered at 845 ºC; D – sample 4 sintered at 860 ºC.

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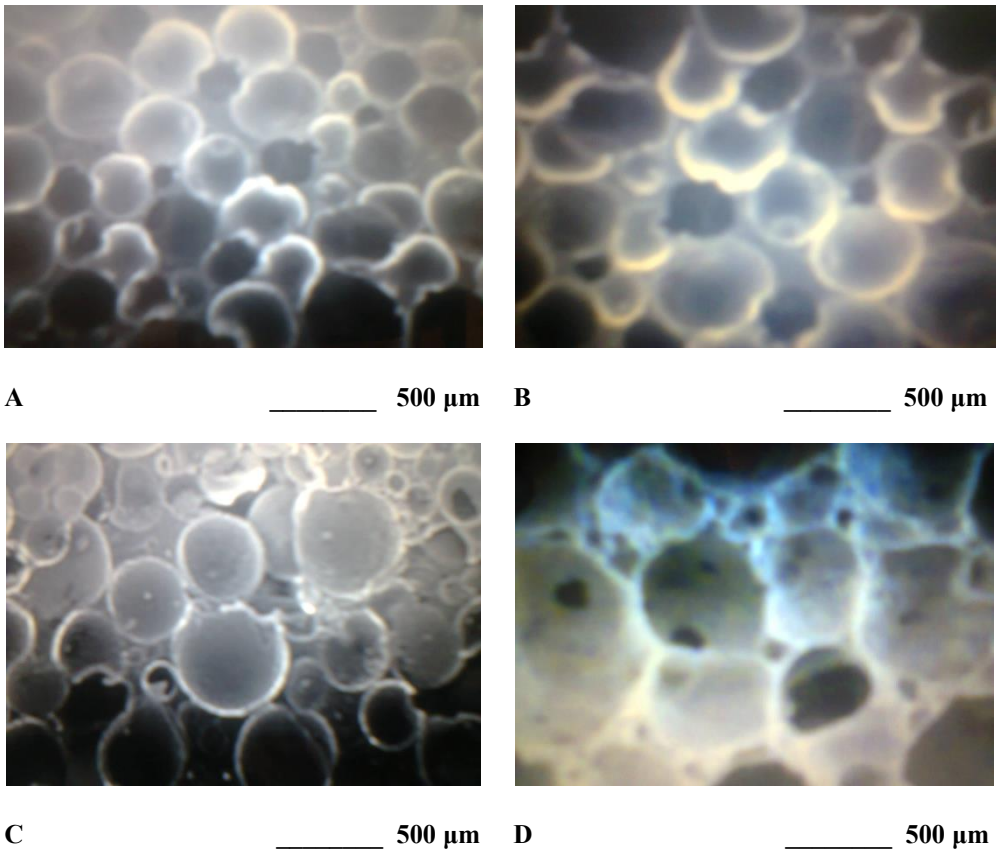

Figure. 3. Microstructural images of the glass foam samples

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

According to Figure 3, the microstructural configuration of glass foam samples was homogeneous, the pore size being below 550 µm in the all experimental variants. The microstructural aspect of the sample corresponding to variant 4 was partially different from the other variants by the coalescence of some cells of the foamed material according to those mentioned above and which influenced the characteristics of this sample.

In order to choose the optimal variant of glass foam manufactured by the unconventional technique of microwave heating, the extreme variants (1 and 4) were excluded, the first because the apparent density and thermal conductivity had too high values negatively influencing the thermal insulation properties of this material and the sample corresponding to variant 4 due to the morphological causes noted above. From samples 2 and 3, sample 2 was chosen as optimal considering its compressive strength (6.1 MPa) much higher than sample 3 (3.2 MPa). By comparison, the values of the apparent density and thermal conductivity of sample 3 were much closer to the values of the same characteristics of sample 2. In addition, the value of the specific energy consumption of sample 2 was lower (0.83 kWh/kg) than the consumption value of sample 3 (0.89 kWh/kg).

A comparison of the experimental results obtained by foaming the flat glass waste with Si_3N_4 as a foaming agent and MnO_2 as an oxygen supplying agent by unconventional and conventional technique, respectively (Llaudis et al., 2009) showed their approximate similarity in terms of quality. Unfortunately, a comparison in terms of energy was not possible due to the lack of this information in the literature. What is still recognized in the literature (Kharissova et al., 2010) is the remarkable energy efficiency of microwave heating processes in general and that the use of industrially designed microwave equipment could improve this efficiency by up

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to 25% compared to the values obtained by the Daily Sourcing & Research company on very low power ovens (0.8 kW).

Conclusion

The objective of the research that underlies the current work was to manufacture a glass foam with a good structural homogeneity from flat glass waste using Si3N4 as a foaming agent and MnO2 as an oxygen supplying agent. The originality of the paper is the application of the unconventional microwave heating technique, faster and more economical, unlike the other papers in the same area published in the literature, followers of the traditional conventional heating technique. Four experimental variants including the glass waste (between 92.5-96.0 wt.%), Si3N4 (kept constant at 2 wt.%), MnO2 (between 2.0-5.5 wt.%) and water addition (kept constant at 14 wt.%) were tested on the 0.8 kW-microwave oven. As the MnO2 content increased, the sintering/foaming temperature increased from 815 to 860 ºC and the heating time also increased from 35 to 44 min. The specific energy consumption was very economical reaching a minimum value of 0.78 kWh/kg for the minimum MnO2 content and the maximum value of 0.98 kWh/kg corresponding to the maximum MnO2 content. The main physical, thermal and mechanical characteristics of glass foam samples were: apparent density between 0.38-0.57 g/cm3, porosity between 72.9-85.6%, thermal conductivity in the range 0.085-0.123 W/m·K and compressive strength between 3.2-7.8 MPa.

At the maximum porosity value (85.6%) corresponding to sample 4, the coalescence of some cells and the formation of interconnection channels in their thicker walls were observed, influencing the characteristics of the sample by increasing the apparent density and the compressive strength. The best sample was considered sample 2. Sintered at 830 ºC, its main physical, mechanical, thermal and morphological characteristics were: apparent density of 0.47 g/cm3, porosity of 77.6%, thermal conductivity of 0.105 W/m·K, compressive strength of 6.1 MPa and pore size between 0.15-0.40 mm. The optimal glass foam sample has the required characteristics of a thermal insulation material usable under mechanical stress conditions in civil engineering.

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