

NONCONVENTIONAL HEATING TECHNIQUE TO PRODUCE GLASS-CERAMIC FOAM FROM GLASS WASTE AND OLD CLAY BRICK WASTE

Marius Florin Dragoescu[1],Lucian Paunescu[2],Sorin Mircea Axinte[3,4] and Ana Casandra Sebe[5]
[1] Daily Sourcing&Research SRL Bucharest, Romania, mar_dmf@yahoo.com
[2]Junkoeko SRL Slobozia, Romania, lucianpaunescu16@gmail.com
[3] Daily Sourcing&Research SRL Bucharest, Romania, sorinaxinte@yahoo.com
[4] Department of AppliedChemistry and Materials Science, University POLITEHNICA of Bucharest, Romania, sorinaxinte@yahoo.com
[5]CosfelActual SRL Bucharest, Romania, casandrasebe@yahoo.com

ABSTRACT: The use of microwaves as a nonconventional energy source in the manufacturing process of glass-ceramic foam is presented in the paper. The main raw materials, representing 88wt.% of the load, are bottle glass waste and old clay brick waste, available in very large quantities. The weight ratio of the two waste types was varied between 3/1 – 3/2, aiming the improvement of the glass-ceramic foam mechanical strength. Substituting 40% from the glass waste mass with clay waste, it was obtained a high compressive strength of 3.35 MPa, in conditions where the apparent density and the thermal conductivity had relative low values (0.78 g/ cm^3 and 0.099 W/ m · K, respectively). Due to his physical, mechanical and morphological features, the foamed product is usable in construction as replacer of similar materials existing on the market.
KEYWORDS: microwave, nonconventional, glass-ceramic foam, glass waste, clay waste, mechanical strength.

1. INTRODUCTION

In the last decades recycling of non-hazardous waste has become a common practice, especially in the world's highly industrialized areas. Numerous types of inorganic non-metallic silicate wastes [1] can be used as raw material to obtain glass-ceramic foams, such as coal ash, metallurgical slags, fly ash and filter dusts from waste incinerators, mud from zinc hydrometallurgy, sludge, glass waste etc. in various combinations [2]. Glass-ceramics have at least one crystalline phase and one amorphous of residual glass [1]. Sintering/ foaming (by the addition of a foaming agent) at high temperature of recycled silicate wastes generates a porous material with a very low thermal conductivity and high or at least acceptable mechanical strength, usable as a building material either for thermal and acoustic insulations or for specific conditions in the case of those with high mechanical strength (aggregate for lightweight concrete, road construction, infrastructure foundation, sport grounds, architectural panels, etc.). Also, the old clay brick as a component of the masonry rubble (together with concrete and mortar from demolition and rehabilitation of buildings) is an aluminosilicate waste, which can be recycled. Both the glass waste and the clay brick waste are suitable in large quantities and have high annual generation rates [3-6]. Several papers presented in the literature indicate researchers' concerns for the use of clay and glass waste in foaming processes, aiming either increasing the mechanical strength of glass-ceramic foam by partial replacement (up to 20 wt.%) of the glass with clay [7] or obtaining a ceramic clay with lower density by partial substitution (up to 50 wt.%) of clay with glass waste [8-11]. It should be noted that all known foaming processes (industrial or experimental) were achieved by conventional heating methods (electrical resistances or fuel consumption).Although known from the middle of the last century as a fast, economical and clean heating technique, the use of microwave energy, as an nonconventional source, has been limited to a few areas of application (of which, the use in household for food preparation is the best known). Also, the vulcanization of rubber and manufacture of polymer-wood composites were application fields of this energy form. Only in recent years it has been experimentally found that the microwave energy could be applied too for other material types (ceramics, polymers, metals, organics, glass etc. [12]. Since 2016 the manufacture of glass foam from glass waste using microwave radiation is being experimentally investigated in the Romanian company Daily Sourcing & Research Bucharest, the results being presented in the literature. The latest results of the Romanian company were experimentally obtained in the manufacturing process of a glass-ceramic foam with superior mechanical strength by heating in microwave field using glass waste and clay brick waste as main raw material.Further, the small-scale microwave manufacturing technology of this product and its physical, mechanical and morphological characteristics are presented.

2. METHODS AND MATERIALS

2.1 Methods

The method adopted for the sintering and foaming of the raw material is that of microwave heating, using a 0.8 kW-domestic microwave oven (Figure 1a) adapted to withstand to the high thermal stress (up to 1200 °C). The microwave heating has a peculiarity that distinguishes it clearly from the conventional heating. The material is first heated into its core and then the heat spreads to the peripheral areas. For this reason, the thermal protection of the material (or of a microwave susceptible crucible containing the material) is required, preferring the ceramic fiber coating (Figure 1b). From the own experience, to reduce the heating rate, which is excessive in the case of the direct heating of silicate materials (over 35 ºC/ min), the use of a ceramic tube made of silicon carbide and silicon nitride with the wall thickness of 3.5 mm was adopted, placed outside the metallic crucible containing the material. Thus, the heating is carried out indirectly through the ceramic wall and the heating rate is reduced below 20 °C/ min, ensuring the proper conditions to obtain a homogeneous porosity material. The thermal control of the heating process is achieved with a radiation pyrometer mounted above the furnace. The upper metallic wall of the oven has a central hole for viewing the surface of the heated material.

a b

Figure 1. The experimental microwave equipment a – the 0.8 kW-microwave oven; b – ceramic tube covered with ceramic fiber.

2.2 Materials

The materials used in the experiments are: old clay brick waste, green bottle glass waste, coal ash and silicon carbide as a foaming agent. The chemical composition of the raw material components is shown in Table 1.

Table 1. Chemical composition of raw material, wt.%

Chemical composition	Raw material		
	Clay [13]	Glass [14]	Coal ash*
SiO_2	56.4	71.8	46.5
Al_2O_3	27.4	1.9	23.7
CaO	1.2	11.8	7.9
Fe_2O_3	7.2	0.01	8.6
MgO	1.4	1.2	3.2
Na_2O	1.0	13.1	6.0
K_2O	4.4	0.1	4.1
Cr_2O_3	-	0.9	-
Other oxides	-	0.01	-

*Based on the own determination.

The waste glass and clay brick waste were mechanically crushed and then ground into an electrically operated device and sieved to a particle size of less than 300 µm. The coal ash purchased from the Paroseni (Romania) thermal power station was sieved to a grain size below 150 µm. Silicon carbide, purchased from the market, had a grain size below 80 µm. The raw materials were adopted due to their composition, which influences the characteristics of the foamed product. Clay contains predominantly SiO_2, Al_2O_3 and to a lesser extent Fe_2O_3. Glass waste contains SiO_2 in very high proportion and Na_2O and CaO, both above 10%. Coal ash has SiO_2 and Al_2O_3 in its composition as main components and also Fe_2O_3, CaO and Na_2O with proportions between 6.0 - 8.6%. Each of the mentioned components of the mixture of raw material has important influences on the foaming process and the characteristics of the final product. Thus, SiO_2 has a reduced thermal expansion and therefore a good thermal shock resistance [15]. Al_2O_3 contributes to increasing the durability of the material and also reduces the melting rate of the glass [16]. Na_2O increases the viscosity of the molten material and CaO favorably influences the crystallization process [15]. It should be noted that Na_2O and K_2O existing in all raw materials favor the microwave field heating due to the correlation between the electrical conductivity of the materials containing these oxides and the absorption of microwaves [17]. Fe_2O_3 is a contaminant in the glass composition, but having microwave susceptibility it favors the heating of the glass at normal intensity starting at the room temperature, despite the consistent presence of the microwave transparent components (SiO_2, Al_2O_3), that are rapidly heated only at higher temperatures [18].

2.3 Characterization of the samples

The samples of the glass-ceramic foam were tested in laboratory to determine the main physical, mechanical and morphological features. Apparent density, porosity, compressive strength, thermal conductivity,water absorption and crystallographic structure were determined using current methods [19-22].

3. RESULTS AND DISCUSSION

The experiments were carried out in fourcompositional variants, comprising mixtures of green bottle glass waste, clay brick waste and coal ash as raw material, silicon carbide as a foaming agent and water addition as a binder. Their weight proportions are shown in Table 2.

Table 2. Chemical composition of raw material, wt.%

Material	Variant			
	1	2	3	4
Green bottle glass	66.0	61.6	57.2	52.8
Clay brick	22.0	26.4	30.8	35.2
Coal ash	9.0	9.0	9.0	9.0
Silicon carbide	3.0	3.0	3.0	3.0
Water addition	20.0	20.0	20.0	20.0

The weight proportions of the basic raw materials (glass waste and clay brick waste) were varied from the 75/ 25 ratio to 60/ 40, while the proportions of coal ash, silicon carbide and water additions have been kept constant at values considered optimal on the basis of the own previous experience.

The main functional parameters of the manufacturing process of glass-ceramic foam in microwave field are presented in Table 3.

Table 3. Functional parameters of the manufacturing process

Parameter	Variant			
	1	2	3	4
Raw material quantity (dry/ wet) (g)	250/ 300	249/ 298.8	250/ 300	248/ 297.6
Sintering/ foaming temperature (ºC)	1000	1025	1040	1060
Heating duration (min)	53	57	60	62
Average heating rate (ºC/ min)	18.5	17.6	17.0	16.8
Glass-ceramic foam quantity (g)	243	241	242	239
Specific consumption of electricity (kWh/ kg)	2.91	3.15	3.31	3.46

Maintaining a relatively constant amount of raw material around 250 g (including also the foaming agent), the temperatures required for sintering and foaming have increased from 1000 °C (corresponding to the lowest clay ratio) to 1060 °C (in the case of the highest ratio). Obviously, the duration of the process has increased from 53 to 62 minutes. The heating rate was maintained in a narrow range between 16.8 - 18.5 C/ min, lower in the case of the low clay ratio. The specific energy consumption varied between 2.91 – 3.46 kWh/ kg, the maximum value corresponding to the process temperature of 1060 ºC. A comparison between the nonconventional heating process presented in the paper and the conventional processes mentioned in the literature can not be achieved due to the absence of data on this functional parameter. In addition to the theoretical considerations that favor the nonconventional process, the heating rates used in the processes described in the literature (between 2 - 10 ºC/ min) lead to significantly higher values of conventional energy consumptions.

The physical, mechanical and morphological features of the glass-ceramic foam samples are shown in Table 4.

Table 4. Physical, mechanical and morphological features of the glass-ceramic foam samples

Feature	Variant			
	1	2	3	4
Apparent density (g/ cm^3)	0.53	0.61	0.70	0.78
Porosity (%)	78.3	74.6	72.0	68.8
Compressive strength (MPa)	1.62	1.91	2.63	3.35
Thermal conductivity (W/ m · K)	0.080	0.085	0.094	0.099
Water absorption (%)	3.7	4.0	3.2	3.1
Pore size (mm)	1 – 4	1 – 3	0.9 – 2.5	1 – 3

According to the data from Table 4, the high ratio of clay in mixture with the glass waste led to the significant increase of the compressive strength of the glass-ceramic foam. If at the glass/ clay weight ratio of 75/ 25 the compressive strength was 1.62 MPa, at the 60/ 40 ratio, this increased to 3.35 MPa. The clay addition has increased the apparent density up to 0.78 g/ cm^3 and, implicitly, the thermal conductivity of the foam up to 0.099 W/ m · K. The water absorption was maintained at relatively low values (between 3.1 – 4.0%), although it is known that the clay is highly water absorbent.

Figure 2 shows images of the cross sections of the glass-ceramic foam samples.

The cross sections of glass-ceramic foam have an uniform pores distribution with the pore size up to 4 mm, indicating that the porous material is suitable for using as a building insulator.

The microstructural analysis of the glass-ceramic foam samples was performed with a Smartphone Digital Microscope. The pores sizes in the cross section (see Table 4) could be determined through this technique. Images of the microstructure of samples 3 and 4 are shown in Figure 3.

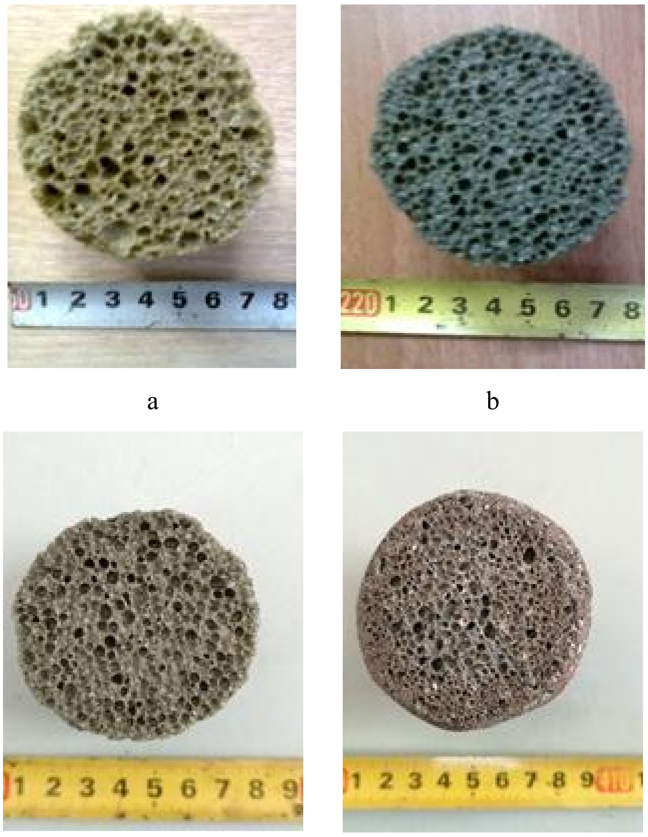

Figure 2.Images of the cross sections of the glass-ceramic foam samples.

a - sample 1; b – sample 2; c – sample 3; d – sample 4

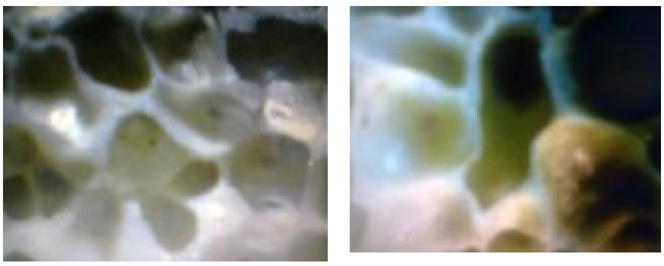

Figure 3.Microstructural images of the cross sections of the glass-ceramic foam samples.
a - sample 3; b – sample 4.

The XRD analysis, performed on a X-ray diffractometer Bruker AXS Advance with CuK α radiation, allowed the identification of the crystalline phases of the sample 4 after the thermal treatment at 1060 ºC. The main crystalline phases were anorthite,cristobalite and quartz.

4. CONCLUSION

Microwave energy as a nonconventional, fast and economical source is used in the manufacturing process of glass-ceramic foam with high mechanical strength, obtained from existing wastes in large quantities, with large annual generating rates: post-consumer bottle glass, clay brick from demolition and rehabilitation of buildings and ash from coal combustion processes in thermal power stations.

The literature presents several works devoted to testing the influence of the proportion of glass and clay in raw material mixtures for the production of high strength porous materials, using conventional heating methods in all cases.

The paper aimed at increasing the mechanical strength of the product using clay waste in a suitable weight ratio, allowing to maintain relatively low apparent density and thermal conductivity and to obtain an usable material as a building insulator.The physical, mechanical and morphological characteristics of the glass-ceramic foam using the glass/ clay ratio of 60/ 40 are: apparent density: 0.78 g/ cm^3, thermal conductivity: 0.099 W/ m · K, compressive strength: 3.35 MPa, water absorption: 3.1%, pore size (uniformly distributed): 1 - 4 mm.

The specific energy consumption for producing the glass-ceramic foam with a glass/ clay ratio of 60/ 40 was 3.46 kWh/ kg. This value, strongly influenced by the low quantity of the foamed material, could not be compared to similar processes that use the conventional heating technique due to the absence of this functional parameter in the literature. The heating rate of 16.8 ºC/ min used in the experiments presented in this paper was much higher compared to the heating rates indicated in the literature (2 - 10 ºC/ min) and this could be an indication of the superior energy efficiency of the nonconventional method.

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