FOAM GLASS WITH LOW APPARENT DENSITY AND THERMAL CONDUCTIVITY PRODUCED BY MICROWAVE HEATING

Marius Florin Dragoescu
Daily Sourcing & Research Bucharest
ROMANIA
mar_dmf@yahoo.com

Sorin Mircea Axinte
University "Politehnica" of Bucharest
ROMANIA
sorinaxinte@yahoo.com

Lucian Paunescu
Daily Sourcing & Research Bucharest
ROMANIA
lucianpaunescu16@gmail.com

Alexandru Fiti
Cosfel Actual Bucharest
ROMANIA
feliss2014@gmail.com

ABSTRACT

The Romanian company Daily Sourcing & Research Bucharest, that has researched in the last time foam glass manufacturing processes by the microwave irradiation method, has been concerned to improve its physical characteristics. Aiming to reduce the apparent density and the thermal conductivity of the foam glass, required for use as insulating material in construction, the processing quality level of glass waste has been significantly increased so its granulation was reduced below 63 µm, compared to 80 – 150 µm used in the previous experiments. Tests carried out on an adapted domestic microwave oven in working conditions similar to those of heating the finely ground and pressed raw material on the conveyor belt of a tunnel furnace, have led to obtaining porous materials with apparent density between 0.15 – 0.19 g/ cm^3 and thermal conductivity in the range 0.034 – 0.040 W/ m·K.

Keywords: Foam glass, microwave, glass waste, foaming, apparent density.

INTRODUCTION

Foam glass is a sintered porous material obtained from a finely ground mixture of glass waste and one of the foaming agents (calcium carbonate, black carbon, graphite, silicon carbide etc.) at high temperatures of over 750 ºC. The foaming mechanism consists in releasing a gas (mainly, carbon dioxide) through a chemical reaction to decompose or oxidize the foaming agent. The sintering temperature of the powder mixture must be correlated with the temperature at which the chemical reaction mentioned above occurs, so that the release of the gas is produced under the conditions of a suitable viscosity of the sintered material. Thus, the gas bubbles that are developed inside its mass are blocked and by the cooling form a porous structure with low apparent density and thermal conductivity and acceptable mechanical strength (Hurley, 2003).

Due to these characteristics, the foam glass is best suited as a rigid insulating material in construction. It has excellent fire resistant properties (unlike the polymeric materials) and it is rodent resistant, sound absorber, non-toxic and non-water absorbent (Hurley, 2003). Also, foam glass is used as floors and wall tiles, architectural panels, filters, absorbers, gas sensors. Foam glass with high compressive strengths can be used as aggregate for lightweight concrete, in road construction, infrastructures foundation, sports grounds (Hurley, 2003; Scarinci, Brusatin & Bernardo, 2005, Rawlings et al., 2006, Paunescu et al., 2017 a).

ISSN 2056-5860

LITERATURE REVIEW

According to the information in the literature (Hurley, 2003), manufacturing technologies of foam glass are currently applied industrially. All technologies are based on conventional heating methods (fossil fuels combustion or electric resistances). The main products industrially made are: "Technopor", produced by Misapor Switzerland Company with branches in Germany, France and Austria and "Foamglas", manufactured by Pittsburgh Corning Company with branches in United States, Europe (Belgium, Czech Republic) and China.
The technical novelty adopted by the company Daily Sourcing & Research in the manufacturing process of foam glass is the use of the microwave energy. The advantage of the microwave using consists in the direct heating only of the material subjected the thermal treatment. This unconventional technique allows a fast, "clean" and economical heating. Though the microwave energy is currently used in the household, this heating system is not yet industrially applied than in an extremely low scale (vulcanization of rubber and manufacture of polymer/ wood composites). Recently, it was demonstrated that the microwave energy can be effectively used in the case of heating for much other material types: organics, ceramics, polymers, metals, composites etc. (Kharissova et al., 2010; Paunescu et al., 2017 b).

The experimental results obtained by the Romanian company, published (Paunescu et al., 2016; Dragoescu et al., 2018) or in process of publishing (Dragoescu et al., 2018 in press) in Romanian or international journals, have shown that, by the microwave heating, the physical, mechanical and morphological characteristics are almost similar to those manufactured by the conventional heating. However, due to the processing conditions of raw material (mainly, glass waste) which did not allow to obtain a smaller granulation of glass waste of 63 – 130 µm, the physical characteristics of foam glass samples were affected, the minimum value of apparent density being 0.25 g/ cm^3 and the corresponding thermal conductivity being 0.041 W/ m·K (Paunescu et al., 2017 b).

METHODOLOGY
The Adopted Method

The adopted method consists of providing the working conditions similar to those of foam glass manufacturing in a tunnel furnace with conveyor belt, the raw material being loaded pressed. The sidewalls and the plate vault covered with a silicon carbide layer as a microwave susceptible material are simulated by the placement with the opening down of a cylindrical silicon carbide crucible with the diameter of 125 mm, the height of 100 mm and the wall thickness of 3.5 mm. It completely covers a metal crucible containing the powder mixture previously pressed with the diameter of 107 mm, the height of 39 mm, the bottom thickness of 6 mm and those of sidewall of 1 mm. The two crucibles are placed in the position described above into a 0.8 kW domestic microwave oven adapted to the working conditions at high temperature. To avoid the heat loss outside, the silicon carbide crucible is covered with two layers of ceramic fiber mattress and the metal crucible is placed on the metal support in rotation, provided with several thick layers of ceramic fiber. The upper area of the thermal protection of the silicon carbide crucible has provided a hole of about 30 mm in the ceramic fiber layers, corresponding to the hole position from the upper wall of the oven housing, so that the silicon carbide surface is visualized with a Pyrovar type radiation pyrometer (measuring field: 600 – 2000 ºC). The temperature of the foamed material was determined through a correlation relationship with the temperature measured on the silicon carbide wall surface.

Figure 1 shows an image of the adapted 0.8 kW domestic microwave oven (a) as well as images of the metal and silicon carbide crucibles (b) and the thermal protection with ceramic fiber of the silicon carbide crucible placed with the opening down (c).

a b c

Fig. 1. The microwave experimental equipment
a – adapted 0.8 kW domestic microwave oven; b – the metal and silicon carbide crucibles; c – thermal protection with ceramic fiber of the silicon carbide crucible placed in the oven with the opening down

The methodology of experimentation consisted in testing the six variants of the mixture composition formed by bottle colorless glass waste and calcium carbonate as foaming agent. The glass waste granulation was below 63 μm, from which 60% represented the grain-size fraction below 32 μm. The mixture was wetted with 8.3% water addition as binder for the pressing process. The powder mixture composition corresponding to the tested variants are shown in Table 1. According to the data from this table, the weight ratio of glass waste had values between 98.0 – 99.0% and the weight ratio of calcium carbonate was between 1.0 – 2.0%. These weight proportion ranges are the result of previous experiments conducted in various other conditions in the company Daily Sourcing & Research. Therefore, the ranges adopted for the test were limited to the values specified above.

Table 1: **Powder mixture composition**

Variant	Bottle colorless glass waste wt.%	Calcium carbonate wt.%	Water addition wt.%
1	99.0	1.0	8.3
2	98.8	1.2	8.3
3	98.6	1.4	8.3
4	98.4	1.6	8.3
5	98.2	1.8	8.3
6	98.0	2.0	8.3

The foam glass samples, resulted after the sintering and foaming experimental process, were tested in laboratory to determine the physical, mechanical and morphological characteristics.

Apparent density, porosity, thermal conductivity, compressive strength, hydrolytic stability and water absorption were determined by the current methods (Paunescu et al., 2017 b; "Manual", 1999; Anovitz & Cole, 2015; "ISO", 1985; "Calculation", 2017).

Materials

As noted above, the experimentally used raw material was bottle colorless glass waste, having the following chemical composition: 71.8% SiO_2, 1.9% Al_2O_3, 12.0% CaO, 1.0% MgO and 13.3% Na_2O (Paunescu, 2017 a). The glass waste was broken and ground in a ball mill, reaching grain size less than 63 µm. The granulation analyzes showed that about 60% represents grain sizes below 32 µm. This advanced processing of the raw material constituted an untapped technological advantage in the own previous experiments aimed at producing foam glass.

Calcium carbonate as foaming agent was used without other mechanical processing such as it was purchased from the market, having a very fine granulation below 40 µm.

After dosing the quantities of the two components, the powder mixture corresponding to the each experimental variant was homogenized together the amount of additional water added (8.3% over the amount of raw material) in a small laboratory installation.

Then, the wet mixture was loaded into a demountable mold and was pressed by mechanical methods up to about 10 – 12 MPa. After removing from the mold, the pressed material was placed freely on the bottom of the metal crucible.

RESULTS

The experiments aimed especially the significant improvement of the main physical characteristics of foam glass (apparent density, porosity, thermal conductivity) manufactured in microwave field from waste glass by reducing the granulation of this raw material. In fact, all own previous experiments in the last two years have used the microwave energy as an unconventional source of heating (fast, "clean" and economical) in the foam glass manufacturing process.

The functional parameters of the process carried out in the microwave oven described above are shown in Table 2.

Table 2: **Parameters of the sintering and foaming process**

Variant	Raw material amount g		Sintering/ Foaming temperature ºC	Average speed ºC/ min		Soaking time in the stopped oven min	Foam glass amount g
	Dry	Wet		Heating	Cooling		
1	319.0	345.5	822	16.5	5.8	30	305.7
2	319.0	345,5	825	16.4	5.1	30	304.9
3	319.0	345.5	829	16.9	5.5	30	306.2
4	319.0	345.5	833	16.0	5.9	30	303.1
5	319.0	345.5	838	15.6	6.0	30	303.7
6	319.0	345.5	839	15.2	5.6	30	301.4

In accordance with Table 2, the raw material amounts (dry and wet) were kept constant: 319 g and, respectively, 345.5 g. The sintering temperature of the powder mixtures, at which the foaming occurred were in the range: 822 – 839 ºC, the lowest temperature corresponding to the variant 1 (99% glass waste and 1% calcium carbonate) and the highest value corresponding to the variant 6 (98% glass waste and 2% calcium carbonate). The average heating speeds are relatively high (between 15.2 – 16.9 ºC/ min) due to the low thickness of silicon carbide crucible, which allows the simultaneous microwave heating of the material (partially direct and partially indirect). All variants within the range of compositions shown in Table 1 have this heating mode feature. The foam glass cooling was carried out slowly with speeds between 5.1 – 6.0 ºC/ min. starting with the cooling into the oven for 30 min. The foam glass amounts had values between 301.4 – 306.2 g, representing 94.5 – 96.0% from the dry raw material.

The physical, mechanical and morphological characteristics of the foam glass samples are shown in Table 3.

Table 3: **Physical, mechanical and morphological characteristics**

Variant	Index of volume growth	Apparent density g/ cm^3	Poro-sity %	Thermal conducti-vity W/ m·K	Compressive strength MPa	Water absorption %	Pore size mm
1	3.40	0.16	92.7	0.034	1.22	1.0	0.6 – 0.9
2	3.50	0.15	93.2	0.035	1.18	1.3	0.8 – 1.1
3	3.40	0.15	93.0	0.035	1.20	0.8	0.7 – 1.4
4	3.30	0.18	91.8	0.038	1.15	0.9	0.9 – 1.5
5	3.50	0.18	91.6	0.037	1.16	1.5	1.0 – 1.7
6	3.60	0.19	91.4	0.040	1.12	1.4	1.0 – 1.9

The main objective of the research was accomplished. Porous products with very low apparent density and thermal conductivities as well as very high porosities were experimentally manufactured. According to the data from Table 3, the apparent density values are in the range 0.15 – 0.19 g/ cm^3, the lowest values corresponding to the variants 1 – 3 (98.6 – 99.0% glass waste and 1.0 – 1.4% calcium carbonate). By the decrease of the glass waste/ calcium carbonate ratio to 98.0/ 2.0, the density value tends to increase slightly, reaching 0.19 g/ cm^3. Closely related to the apparent density of the material, the thermal conductivity has also very low values between 0.034 – 0.040 W/ m·K, the values increasing from the variant 1 to the variant 6. The foam glass porosity reaches the highest values in the case of the variants 1 – 3, between 92.7 – 93.2%, being directly influenced by the low values of the apparent density. The compressive strength of these glass foams falls within the normal range (1.12 – 1.22 MPa) of a material characterized by such low density values, being considered acceptable for an insulating material. Also, the amount of water absorbed by the glass foams is almost negligible (0.8 – 1.5%), being practically non-absorbing to water, such as the glass foams currently manufactured by conventional heating methods.

ISSN 2056-5860

The longitudinal sections through the foam glass samples (shown in Figure 2) are characterized by homogeneous pores matrices. The calcium carbonate weight proportion in the mixture composition influenced the pores size, at lower proportions the sample macrostructure being finer (pores size around 1 mm) and at weight proportions of 1.8 – 2.0% the pores size exceeding 1 mm up to 1.9 mm (see Table 3).

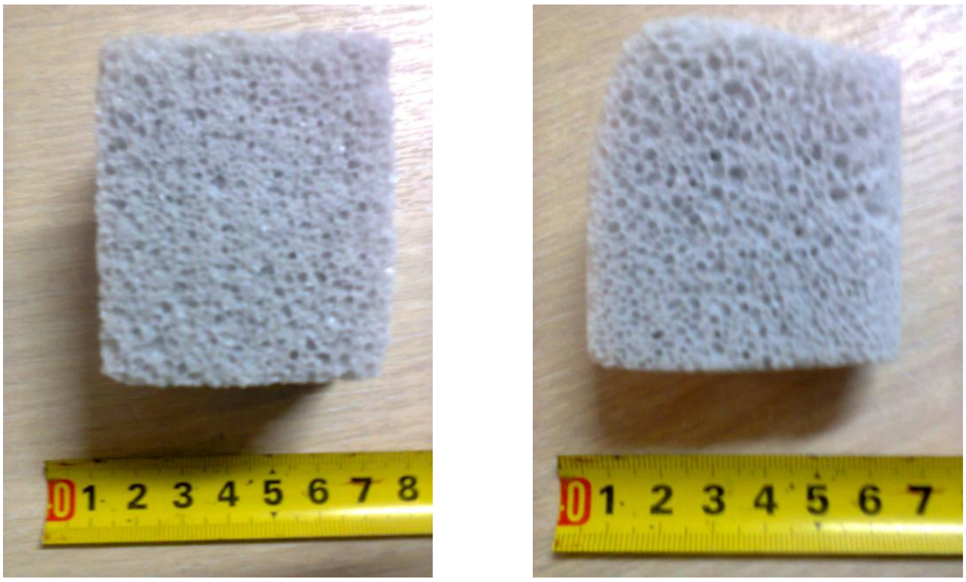
a

b

c

d

ISSN 2056-5860



e f

Fig. 2 Images of the longitudinal section of the samples
a – sample 1; b – sample 2; c – sample 3; d – sample 4;
e – sample 5; f – sample 6.

The tests for determining the hydrolytic stability of samples, using 0.15 ml of 0.01M HCl solution to neutralize the extracted Na_2O, showed that the stability joins in the hydrolytic class 2, the extracted Na_2O equivalent being in the range 31 - 57 µg.

DISCUSSION

The microwave heating of the powder mixture based on glass waste for manufacturing the foam glass has been previously researched by the Romanian company Daily Sourcing & Research. The experimental results showed that the thermal process begins inside the sample, where the temperature increases rapidly, influenced by the microwave irradiation. The foaming process is developed rapidly from inside to the peripheral area of the sample. This heating mode is not suitable for the materials based on glass because the very high heating speed generates major imbalances in the structure of the material (non-homogeneous structure characterized by high pore size, sometimes even containing very large goals.

From this raison, a solution for diminishing the effect of the microwave field on the material subjected to heating was adopted by placing a wall made of a microwave susceptible material in the way of microwave propagation. Silicon carbide was adopted and the wall thickness was determined experimentally at 3.5 mm in conditions of a relatively large area of the powder mixture. In this way, the electromagnetic waves are partially absorbed in the mass of susceptible material, which is rapidly heated and its inner surface seen by the material transfers heat through thermal radiation, that contributes to the heating process. Also, some of the microwaves penetrate completely the susceptible material and irradiate the sample based on glass waste. Its heating is started from the inside to the outside. Thus, two high temperature centers that act in two opposite directions are formed. The heating speed is sufficiently diminished from over 26 ºC/ min to about 15 – 17 ºC/ min and the effect is favorable for the foaming process and the homogeneity of the porous material macrostructure.

CONCLUSIONS

The objective of the research was to manufacture in microwave field of the glass foam with very low apparent density and thermal conductivity suitable for use as insulating material in construction.

The main solution was improving the glass waste processing and significant reducing its grain-size below 63 μm.

The technology, the equipment and the weight proportion of the mixture components were adopted based on the experience and the previous experimental results of the Romanian company Daily Sourcing & Research in the field of producing foam glass with the microwave energy.

The working technique used in experiments was the placement of the pressed powder mixture on a large metal crucible covered with a cylindrical silicon carbide crucible with thickness of 3.5 mm. The two crucibles were introduced into an adapted 0.8 kW domestic microwave oven. The raw material was colorless bottle glass waste (between 98.0 – 99.0 wt.%) and the foaming agent was calcium carbonate (between 1.0 – 2.0 wt.%), the powder mixture being wetted with 8.3 wt.% water addition.

The experimental results confirmed the obtaining porous products with the following characteristics: apparent density 0.15 – 0.19 g/ cm^3, porosity 91.4 – 93.2%, thermal conductivity 0.034 – 0.040, compressive strength 1.12 – 1.22 MPa, water absorption 0.8 – 1.5%, pore size 0.6 – 1.9 mm.

The products are suitable for using as insulating material in construction.

REFERENCES

Anovitz, L. M. & Cole, D. R. (2015) Characterization and analysis of porosity and pore structures, *Reviews in Mineralogy & Geochemistry*, *80*, 61 – 164.

Calculation of the chemical durability (hydrolytic class, corrosion) of glass (2017) [Accessed 16th May 2016] Available from World Wide Web: http://glassproperties.com/chemicaldurability/

Dragoescu, M. F., Paunescu, L., Axinte, S. M. & Fiti, A. (2018) Influence of the color of bottle glass waste on the characteristics of foam glass produced in microwave field, *International Journal of Science and Engineering Investigations*, *7*, *72*, 95 – 100.

Dragoescu, M. F., Paunescu, L., Axinte, S. M. and Fiti, A. (2018 in press) Simulation foam glass production in a tunnel furnace powered with microwaves, *International Journal of Innovative Science and Research Technology*.

Hurley, J. (2003) *Glass - Research and Development, Final report, A UK market survey for foam glass.* Banbury, Oxon, Great Britain: The Waste and Resources Action Programme Publication.

ISO 719: 1985 (reviewed and confirmed in 2011) Glass – Hydrolytic resistance of glass grain at 98 ºC – Method of test and classification.

Kharissova, O., Kharissov, B. I. & Ruiz Valdés, J. J. (2010) Review: The use of microwave irradiation in the processing of glasses and their composites, *Industrial & Engineering Chemistry Research*, *49*, *4*, 1457 – 1466.

Manual of weighing applications, Part 1, Density (1999) [Accessed 15th October 2016] Available from World Wide Web: http://www.deu.ie/sites/default/files/mechanicalengineering/pdf/manuals/DensityDeterminationmanualpdf

Paunescu, L., Grigoras, B. T., Dragoescu, M. F., Axinte. S. M. & Fiti, A. (2017 a) Foam glass produced by microwave heating technique, *Bulletin of Romanian Chemistry Engineering Society, 4, 1*, 98 – 108.

Paunescu, L., Axinte, S. M., Grigoras, B. T., M. F. Dragoescu & Fiti, A. (2017 b) Testing the use of microwave energy to produce foam glass, *European Journal of Engineering and Technology (EJET)*, 5, 4, 8 – 17.

Paunescu, L., Dragoescu. M. F. & Axinte, S. M. (2016) Comparative analysis of the own experimental techniques of producing the foamed glass-ceramic, *Journal of Engineering Studies and Research, 22, 2*, 55 – 64.

Rawlings, R. D., Wu, J. P. & Boccaccini, A. R. (2006) Glass-ceramics: Their production from wastes - A review, *Journal of Materials Science, 41, 3*, 733-761

Scarinci, G., Brusatin, G, & Bernardo, E. (2005) Glass foams (Chapter 2.7). In Scheffler, M. & Colombo, P., *Cellular ceramics: structure, manufacturing, properties and applications (pp. 158 – 176)*, Weinheim, Germany: Wiley – VCH Verlag GmbH & Co. KGaA.