TESTING THE USE OF MICROWAVE ENERGY TO PRODUCE FOAM GLASS

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ABSTRACT

The paper presents experimental results obtained lately by a Romanian team of researchers in the field of producing foam glass as a building insulation material from very high weight proportion of waste glass (over 98%) and calcium carbonate as a foaming agent at 821 – 830 ºC, in conditions of using the microwave energy. Unlike the conventional heating methods well-known worldwide, the microwave heating of waste glass starting from the ambient temperature has been successfully tested both in an adapted 0.8 kW domestic microwave oven and in an existing microwave reactor with the installed power of 3 kW. The foam glass has the physical and mechanical features (apparent density, porosity, compressive strength, thermal conductivity etc.) comparable to similar products obtained by conventional heating methods, being mainly destined to the building sector.

Keywords: Foam glass, microwave, waste glass.

INTRODUCTION

Foam glass is a porous material obtained by a thermal treatment at high temperature (700 – 1100 ºC) of waste glass with addition of a foaming agent. The foamed material contains numerous evenly distributed bubbles, that give it low density and thermal conductivity. Other characteristics of foam glass as high mechanical strength, non-flammability, thermal stability and high chemical durability recommend this material as a very good insulator, being competing with insulating polymeric materials (M. Scheffler, 2005).

Currently, raw material heating is carried out by conventional methods (fuel burning or electric resistance). Although the advantages of the microwave heating are well-known, this heating type is used on large scale only to the food preparation, 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, glass, sol-gel, metals and composites (O.V. Kharissova, 2010).

Lately, the Romanian company Daily Sourcing & Research Bucharest tested in its experimental basis several variants of foam glass manufacture using the microwave energy, both on an adapted 0.8 kW domestic microwave oven and on an existing microwave reactor with installed power of 3 kW, previously designed and achieved for melting processes of metals. Different foaming agent types (silicon carbide, calcium carbonate, graphite, glycerine), raw material and addition material compositions, material nature and wall thickness of crucibles containing powder mixture of raw material (silicon carbide with thick or thin wall, stainless steel), as well as different processing technologies (heating and cooling speed, soaking times at the foaming temperature) were tested recently, the results being presented in works in process of publishing in Romanian journals.

LITERATURE REVIEW

The manufacture process of foam glass using conventional heating methods is presented in literature in the different technological variants. Very various types of silicate wastes (coal ash, oil shale ash, metallurgical slag, wastes of zinc hydrometallurgy, waste quartz sand) mixed with different foaming agents (silicon carbide, calcium carbonate, black carbon, graphite, coal dust, coal gangue, banana leaves) and mineral additions (sodium silicate, borax etc.) led to producing foam glass with very different physical and mechanical features and implicitly with wide applicability depending on these characteristics (R. D. Rawlings, 2006; Z. Li, 2016; S. Arcaro, 2016). The wide applications refer to foam glasses with high mechanical strength, currently manufactured industrially. The main products are: "Technopor" made by Misapor Switzerland Company with branches in Germany, France and Austria and "Foamglas" made by Pittsburgh Corning Company (USA) with branches in USA and Europe (Belgium, Czech Republic) and China (J. Hurley, 2003).

By comparison, the information on manufacturing methods of foam glass using the microwave energy is very limited. Theoretically, the direct microwave heating involves the susceptible character of the powder mixture of raw material preponderantly consisting from waste glass. A work on the use of microwave radiation for processing the glass (M. Knox, 1997) concludes that the commercial glass is not microwave susceptible up to 500 ºC and so, practically, the manufacture process of foam glass can't be completely achieved using the microwave energy. An additional conventional heating up to 500 ºC is required, complicating the power supply system. Taking into account this conclusion, a market survey for the foam glass manufactured in Great Britain (J. Hurley, 2003) considers that the use of microwave radiation heating in this technological process is probably not appropriate, mainly for the reason of the cost of modifying existing ovens.

M. Knox's theory is not confirmed by the experiments carried out by the Romanian team of researchers, which highlighted that both the usual commercial glass and the powder mixture of waste glass (over 98 wt.%) and calcium carbonate as a foaming agent are susceptible starting from the ambient temperature (L. Paunescu, 2017).

A review on the use of microwave irradiation in the processing of glasses and their composites (O. V. Kharissova, 2010) presents a synthetic situation of industrial application in the field mentioned above of this advanced technique. Thus, the microwave energy is used on large scale in food preparation, vulcanization of rubber and manufacture of polymer/ wood composites. More recent research showed that the applicability field can be extended to organics, ceramics, polymers, glass, metals etc.

Currently, the glass plants use the conventional heating, the microwave energy being applied as an energy source only in special cases as glass reinforced with metals, coating with films, manufacture of some glass-ceramics and vitrification of radioactive waste. According to O. V. Kharissova's work the use of microwave in an industrial oven is much more effective compared to a domestic oven with low power (700 – 1300 W). The industrial oven can be designed as a continuous system with much higher power and higher yield. The industrial equipment allows a high-energy power source with long life, the use of an unique internal protection feature, uniform exposure in the microwave field, much lower process costs.

The literature does not provide data on industrial installations for the production of foam glass or experimental results of laboratory research. An American patent (Y. Matsubara, 1989) describes a method of indirect microwave heating of materials or products, which are moved at low speed along a tunnel furnace. Coatings with carbon dust and silicon carbide as microwave susceptible materials are made on the inner surfaces of the side walls of the furnace. Under the action of microwave radiation, the layers of the susceptible materials mentioned above are heated quickly and intensely and transfer their heat by thermal radiation to the material.

METHODOLOGY

Methodology for Experimental Determining the Microwave Susceptible Character of the Powder Mixture Based on Waste Glass

Previous research carried out by M. Knox and G. Copley (M. Knox, 1997) observed that the usual commercial glass becomes microwave susceptible only at temperatures over 500 ºC and that after about 900 ºC this feature loses its intensity. Therefore, preliminary tests on the influence of microwave radiation on the powder mixture based on waste glass (containing over 98 wt.% waste glass and below 2 wt.% calcium carbonate as a foaming agent) have been carried out on samples obtained by pressing, moistened with 8 wt.% water addition. The experiments have been conducted both on an adapted 0.8 kW domestic microwave oven (Figure 1) and on a 3 kW microwave reactor (Figure 2) existing in the experimental basis, powered with microwaves through the side cylindrical wall. In the both cases, the pressed sample has been thermal protected with ceramic fiber to reduce the heat loss outside the system.

Fig. 1 Adapted 0.8 kW domestic microwave oven

Fig. 2 The 3 kW microwave reactor

The test carried out on the adapted domestic microwave oven allowed visual observation of the process, but the temperature measurement was deficient due to the rotation of the support containing the sample. The second test type allowed the continuous monitoring of the

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temperature of the upper area of the pressed material and, simultaneously, through a mirror, the evolution of the foaming process could be visual observed. The particularity of the heating in the central and lower areas of the sample were not known. After the end of the heating process and the cooling of samples, the effects on their internal morphology could be examined.

Methodology for Experimental Determining the Functional Parameters of the Foaming Process and the Physical, Mechanical and Morphological Features of the Samples

Following the own previous experimental data presented in works in process of publishing in Romanian journals, the use of calcium carbonate as a foaming agent in the manufacture process of foam glass in microwave field resulted to constitute one of the most economical available solutions due to at least two reasons: the relatively low temperature at which the foaming process takes place (a little over 800 ºC) and the very high weight proportion of recycling waste glass which can be processed (over 98%). For this reason the experiments have been directed to this goal.

The chemical composition of waste glass used in experiments (usual commercial bottle colorless glass) is: 71.8% SiO_2, 1.9% Al_2O_3, 12.0% CaO, 1% MgO and 13.3% Na_2O.

The main equipment used to conduct the experiments has been an existing microwave reactor powered by three magnetrons placed equidistantly on the side wall of the reactor cavity (see Figure 2). The installed power is 3 kW.

Previous tests used several microwave heating ways of the material subjected to the foaming process: direct, predominantly direct (partially indirect) and respectively indirect. These heating types have been obtained by the variation of the sample positioning mode in the microwave propagation trajectory. Thus, the pressed material positioning inside the reactor cavity without providing a crucible or a cylindrical wall made of a microwave susceptible material placed between the electromagnetic wave emission area and material creates the conditions to achieve a direct heating. The use of a crucible containing the material or a cylindrical wall placed on the inner edge of the reactor cavity, the both having a thin wall (1 – 2.5 mm) allows the predominant passage of microwave up to the contact with the material and, simultaneously, in a low proportion, the wave absorption into the susceptible material, its heating and the indirect heat transfer by thermal radiation. This heating type is one mixed, predominantly direct. The last heating type is the indirect, obtained by placing a thick wall (15 – 20 mm) made of a susceptible material on the inner edge of the cavity or the use of a crucible also with thick wall containing the powder material. Obviously, the each heating type mentioned above is characterized by a heating speed range, which influences decisively the foaming process and the quality of the obtained porous material. In particularly, in the case of the used experimental reactor, the heating speed could be modified by the variation of the heat loss value outside the system by the thermal protection (with ceramic fiber) of the outer side wall of the reactor cavity. Generally, during the different experiments, the average heating speed had values in the following ranges: 21 – 29 ºC/ min in the case of the direct heating, 8 – 13 ºC/ min in the case of the indirect heating and 16 – 22 ºC/ min in the case of the mixed heating (predominantly direct and partially indirect heating). The best physical, mechanical and morphological features of foam glass were obtained with a 20 mm silicon carbide cylindrical wall placed inside the reactor cavity, which ensures an indirect microwave heating, the powder material being loaded into a stainless steel crucible with the wall thickness of 2.5 mm.

The experiments presented in the current paper have been conducted using the indirect microwave heating method described above. The novelty of this set of experiments, compared to the previous ones, consists in the significant diminishing the grain size of waste glass (by the improvement of the recycling glass processing) from 80 – 150 µm up to 32 – 130 µm.

The heating process in the microwave reactor has been monitored with a pyrometer Pyrovar type (measuring field: 700 – 2000 ºC) mounted above the reactor in front of the viewing hole of its metal cover. The functional parameters of the manufacturing process of foam glass (foaming temperature, duration of the process, heating and cooling speeds, soaking time into the reactor, index of volume growth, consumption of electricity, amounts of raw material and foam glass) were noted for each experiment. The final time of the process was determined both by measuring the material temperature through the orifice of the crucible cover, the sign of foaming being a slight decrease of the temperature and by the visual observation with a mirror.

The foam glass samples, resulted after the sintering and foaming processes were tested in laboratory to determine the physical, mechanical and morphological characteristics. The apparent density was determined by the gravimetric method with the picnometer (Manual, 1999). The porosity was calculated by the comparison method of true and apparent densities of the material, experimentally measured (L. M. Anovitz, 2015). The volumetric proportion of the water absorption was determined by the method of water immersion of the sample (for 48 hours). Determining the thermal conductivity was performed by the guarded-comparative-longitudinal heat flow technique, according to ASTM E 1225 – 04. The compressive strength was measured with an uniaxial hydraulic press. The hydrolytic stability of the samples was measured by the standard procedure ISO 719:1985 with a 0.01M HCl solution (Method, 2011; Calculation, 2017).

RESULTS

Experimental Determining the Microwave Susceptible Character of the Powder Mixture Based on Waste Glass

The first behaviour testing the mixture based on waste glass directly microwave irradiated has been carried out on the 0.8 kW domestic microwave oven. This allowed the visual observation of the way of developing the high temperature front of the material placed freely on the wave propagation trajectory. According to the image in Figure 3, the heating of the powder mixture is initiated inside the material, the heat being transferred to its peripheral areas by thermal conductivity. The photographic image captures the moment when the front of high temperature (over 800 ºC) penetrates the much cooler upper surface of the sample and expands to its side wall, which has also a much lower temperature compared to the hot middle of the sample. The duration of the heating process described above of about 35 min, in the conditions in which the initial mass of the sample was 130 g. The foaming of material has occurred violently, the average heating speed being 23 – 25ºC/ min, but it has been estimated that the maximum process speed exceeded 40ºC/ min. In these inappropriate conditions to achieve a homogeneous foaming as in the case of the conventional heating, the product is empty in its central area on more than 80% of the expanded sample volume. The peripheral areas, which contain pores of acceptable size, are vitrified, hard and have an apparent density close to that of glass, of over 2 g/ cm^3.
The test carried out on the microwave reactor (Figure 2) monitored from a thermal point of view the process. The material temperature measured with the pyrometer through the orifice of the metal cover of the crucible has provided accurate data up to the end of the process, when the expanded material from the sample head climbed up to the level of the metal cover and began to

cool. The heating speed had values comparable to those calculated for the first test. Figure 4 shows an image of a longitudinal section of the foamed product.

Fig. 3 The direct microwave heating test of the powder mixture based on waste glass on the adapted domestic microwave oven

Fig. 4 Internal morphology of the sample directly heated on the microwave reactor

The main conclusions of the tests described above are that the microwave based on waste glass has the microwave susceptible character starting to the ambient temperature, but the use of the direct microwave heating of this material is not suitable to produce foam glass with the features required for the insulating materials destined to the building sector.

Experimental Results of Foam Glass Manufacture Using the Microwave Energy

According to the adopted methodology for determining the functional parameters of the foaming process and the physical, mechanical and morphological features of the foam glass, four variants of producing this porous material from waste bottle glass and calcium carbonate have been tested on the microwave reactor in the Romanian company Daily Sourcing & Research. The weight proportions of waste glass were 98.0; 98.5; 99.0 and 99.2%, corresponding to the variants 1 – 4 in this order. The foaming agent (calcium carbonate) has been used in weight proportions between 0.8 – 2.0%. Waste glass with the grain size in the range 32 – 130 μm, calcium carbonate (below 40 μm) and 8% water addition were been mixed, homogenized and pressed into a stainless steel crucible introduced in a silicon carbide cylindrical tube with the wall thickness of 20 mm mounted in the reactor cavity.

The experimental results are presented in Table 1 and 2.

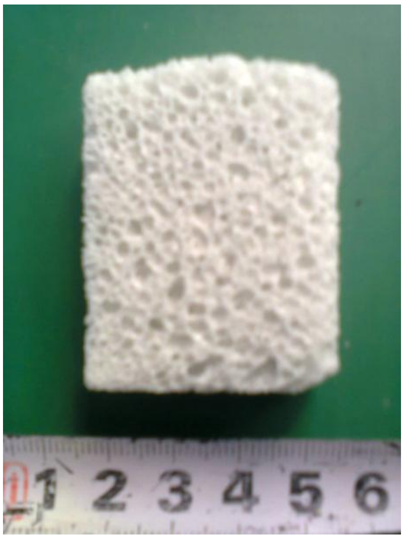
Table 1: Parameters of the sintering and foaming process

Variant	Raw material/ foam glass amount (g)	Sintering and foaming temperature (ºC)	Average speed, (ºC/ min) Heating	Average speed, (ºC/ min) Cooling	Soaking time into the stopped reactor (min)	Specific consumption of electricity (kWh/ kg)
1	117.0 /112.7	830	8.8	7.3	60	10.5
2	117.0/ 111.9	823	9.7	5.7	60	9.9
3	117.0/ 111.0	821	9.5	6.0	60	9.8
4	117.0/ 110.2	830	9.3	6.8	60	10.4

Table 2: Physical, mechanical and morphological characteristics

Var.	Index of volume growth	Apparent density (g/ cm^3)	Porosity (%)	Compressive strength (MPa)	Thermal conductivity (W/m·K)	Water absorption (%)	Pore size (mm)
1	2.40	0.34	84.5	2.0	0.065	7.5	1.0 – 2.5
2	2.15	0.27	87.7	1.6	0.047	6.1	1.0 – 2.0
3	1.95	0.25	88.6	1.3	0.041	5.7	0.7 – 1.3
4	1.90	0.28	87.3	1.4	0.052	4.9	0.4 – 1.0

Longitudinal section of the foam glass samples are shown in Figure 5.



Sample 1

Sample 2

Sample 3

Sample 4

Fig. 5 Images of the longitudinal section of the samples

Images of the pores structure of the four samples were obtained with a macro-epidiascope with image analyzer, being shown in figure 6.

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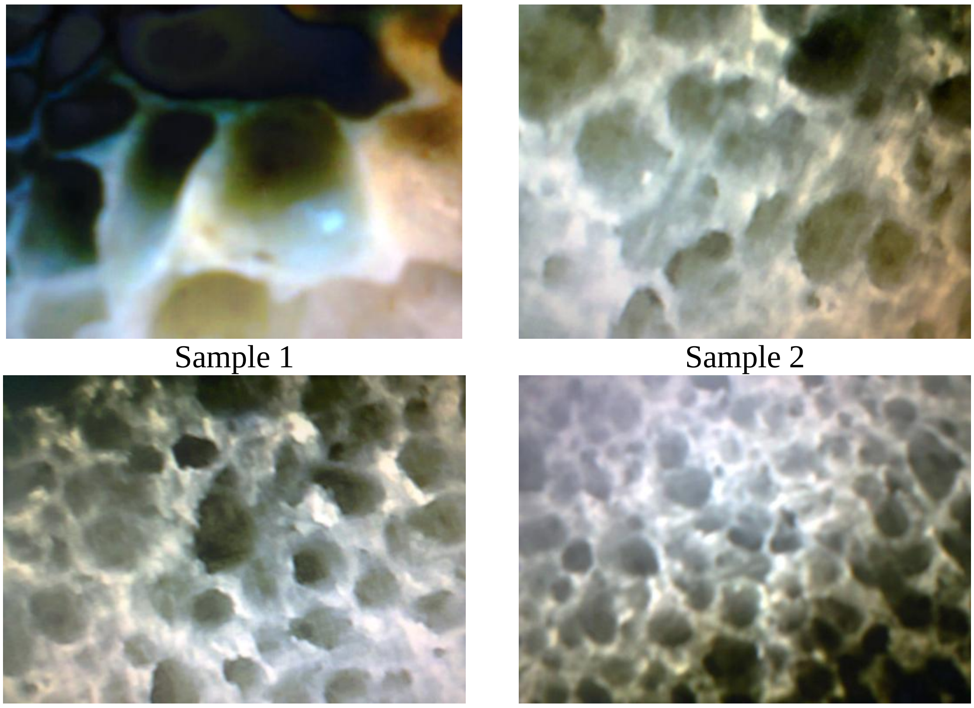

Sample 1 Sample 2

Sample 3 Sample 4

Fig. 6 Images of pores structure determined with a macro-epidiascope with image analyzer

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 30 - 51 µg.

DISCUSSION

The powder mixture consisting in waste glass (over 98%) and calcium carbonate (below 2%) moistened with water (8%) represents certainly an microwave absorbent material between the ambient temperature and below 900 ºC, fact experimentally demonstrated. A theory on the microwave absorption of materials (V. M. Kenkre, 1992) involves absorbing entities such as a vacancies, bivacancies or interstitials which overcome an energy barrier to absorb microwaves to a significant extent.

There is a strong correlation between the electrical conductivity of glasses and the microwave absorption. The glasses with high concentration of alkali can be heated effectively with microwave (U. Kolberg, 2001). The usually commercial colorless glasses used in the current experiment contain 13.3 wt.% and correspond to this requirement.

According to the data from tables 1 and 2, constant amounts of dry raw material (117.0 g) have been used in all four variants, resulting 110.2 – 112.7 g of foam glass, the minimum limit being obtained at the highest ratio waste glass/ foaming agent. Based on the own previous experience, the adopted heating speeds were reduced below 10 ºC/ min (8.8 – 9.7 ºC/ min) to allow the homogeneous development of the technological process. Because the reactor construction does not allow the heat accumulation into a massive refractory structure, like the conventional ovens, the cooling of the porous material has been carried out mainly in the stopped reactor (about 60 min), the average cooling speed being maintained in the range 5.7 – 7.3 ºC/ min.

Unlike other foaming agents, calcium carbonate has tendency of high expanding of the porous material, the index of volume growth reaching 2.40 in the case of 2 wt.% calcium carbonate and 1.90 in the case of a proportion of calcium carbonate of only 0.8 wt.%.

A very important aspect of the advanced processing of raw material, especially of waste glass (32 – 130 µm) constitutes the possibility of diminishing the apparent density of foam glass up to 0.25 g/ cm^3 (for 1 wt.% calcium carbonate – variant 3). Implicitly, the porosity reaches very high values of 88.6% influencing favourable the thermal conductivity of material (0.041 W/ m·K). This feature of foam glass is determinant for the quality of the material used as thermal insulating in construction. Other important mechanical feature of foam glass is the compressive strength. The weight proportion of calcium carbonate of 1%, leads to a lower value (1.3 MPa) compared to the compressive strength values corresponding to calcium carbonate proportions of 1.5 – 2.0%.

The aspect of pores distribution in the structure of foam glass samples is obviously influenced by the weight proportion of the foaming agent. In all variants the pores distribution is relatively homogeneous, differing by the range of pores size, also characteristic in the case of the similar porous materials obtained by the conventional heating (N. Stiti, 2011). In the case of variant 3, with the lowest apparent density of the material, pores size is in the range 700 µm – 1.3 mm.

Referring to the specific consumption of electricity (9.8 – 10.5 kWh/ kg) of the manufacturing process of foam glass in the experimental microwave reactor, in a discontinuous operation regime and having high heat losses outside the system and implicitly a very low energy efficiency, this does not constitute a significant functional parameter. In her work, Oxana Kharissova remarks that an industrial equipment, in continuous regime, allows a high-energy power source, the use of an unique internal protection feature, an uniform exposure to microwave and process costs much lower (O. V. Kharissova, 2010).

CONCLUSIONS

The aim of the research presented in the paper is to obtain a foam glass with high porosity and, simultaneously, with a good compressive strength, features required to the insulating materials for construction.

Unlike the own previous experiments, the method of processing raw material, especially waste glass, was significant improved, its granulation being reduced from 80 – 150 µn to 32 – 130 µm. The experiments have been carried out in the company Daily Sourcing & Research Bucharest on an existing 3 kW microwave reactor.

Preliminary tests have shown that the mixture based on the commercial glass waste is microwave susceptible starting from the ambient temperature, allowing its direct heating.
The optimal solution experimentally determined consists in the indirect heating of raw material, being necessary much lower heating speeds (8 – 13 ºC/ min) than those obtained by the direct heating (21 – 24 ºC/ min).

Foam glass produced with the microwave energy using waste glass (over 98 wt.%) and calcium carbonate (below 2 wt.%) has physical, mechanical and morphological features comparable to those of the similar products obtained by the conventional heating.

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From the point of view of the features required of an insulating material, especially used in construction, the best tested variant is the one obtained from 99 wt.% waste glass and 1 wt.% calcium carbonate in a fine ground powder mixture, homogenized and moistened with 8 wt.% water addition, which allowed to made a porous product with a very high porosity (88.6%), low apparent density (0.25 g/ cm^3) and thermal conductivity (0.041 W/ m·K) and an acceptable compressive strength (1.3 MPa).

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