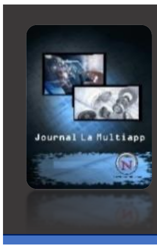

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New Manufacturing Method of Glass Foam by Cold Expansion of Glass Waste

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Abstract

An innovation cold manufacturing method of glass foams is presented in the paper. Traditional foaming agents used in conventional expansion processes of glass waste at high temperature were substituted with aluminium powder in aqueous solution of calcium hydroxide, which releases hydrogen forming gas bubbles in the viscous sludge and then, by solidification, a porous structure typical for the glass foam. The manufactured foam is adequate for using as a thermal insulation material for inner wall of buildings, having the apparent density of 0.31 g·cm-3, the thermal conductivity of 0.070 W/m·K and the compressive strength of 1.32 MPa. The process originality is the use of recycled aluminum waste, melted by an own microwave heating technique and sprayed with nitrogen jets. The process effectiveness is remarkable in economical and energy terms.

Introduction

The trend of recycling materials (plastic, metal, paper and cardboard, glass, rubber, etc.) felt worldwide since the last decades of the 20th century had objective causes: the oil crisis and the major environmental problems due to increasing greenhouse gas emissions affecting the ozone layer of the planet. On the one hand, the oil crisis has led to the reuse in industrial production of waste as raw materials reducing the primary energy consumption. On the other hand, removing the landfills, whose annual generation rate is constantly increasing, has beneficial effects on environmental health.

Large quantities of glass waste are available worldwide for recycling. They come primarily from post-consumer packaging bottles and shards of window glass (flat glass). In 2018, the glass industry recycled 21 % of the total amount of manufactured glass. The recycling rate of the packaging bottle was 32 % and the recycling rate of flat glass was 11 % (Key figures, 2020). The main countries where the glass recycling has a very high rate are Sweden (over 95%), Germany and Italy (Harder, 2018).

However, recycling the glass waste for use as a raw material in the industrial manufacture of new glass involves operations of waste sorting by color, which are expensive (Bourguignon, 2015). For this reason, new fields of application of glass waste have been found, which do not require their qualitative selection before the manufacturing processes. The so-called glass foam, patented and produced since the mid-20th century, uses glass waste as a raw material,

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a foaming agent (inorganic or organic) and, as appropriate, other mineral additives that facilitate the foaming process. The technological process occurs at high temperatures (750-1150 ºC) at which the powder mixture sintered and the foaming agent releases a gas/gaseous compound which forms gas bubbles in the mass with adequate viscosity of the glass. The cooling after stopping the heating process leads to the formation of a porous structure specific to the glass foam.

The properties of glass foam are remarkable. This porous product is light, waterproof, fireproof, anti-corrosive, non-deformable, non-toxic, resistant to rodents, bacteria, insects and acids and has a satisfactory mechanical strength (Scarinci et al., 2005). Due to these simultaneous characteristics, the glass foam can be used as a light thermal insulation material with moderate mechanical strength on the inner walls of the building. Also, it can be used under conditions of mechanical stress in the road and railway constructions, insulation in the perimeter of the building, foundations, roof gardens, drainages, sports fields, thermal insulation of underground energy pipes and storage tanks, etc. (Paunescu et al., 2019; Paunescu & Paunescu, 2020), replacing traditional building materials.

The industrial manufacture of glass foam in all manufacturing companies in the world (Misapor, Pittsburg Corning, Geocell, Glapor, etc.) is carried out by conventional heating techniques (electrical resistances or burning gaseous fuels). The specific energy consumption, in general, is provided in the literature only as an exception. According to Hurley's market study (Hurley, 2003), the Misapor Company has an average energy consumption for the heating process of 100 kWh/m3 (i.e. up to 0.83 kWh/kg), being added an additional 25 kWh/m3 for processing the raw material before loading it in the oven. Pittsburg Corning Company reports an average total consumption of 4.24 kWh/kg, but it also includes material processing, including melting glass waste to correct its chemical composition. The Geocell company reports for the factory in Debrecen (Hungary) an average energy consumption of 140 kWh/m3 (Foam glass, 2014).

Experiments carried out by some authors of the current work in the Daily Sourcing & Research Company aimed at using a microwave heating technique, faster and more economical, for the experimental small-scale manufacture of different types of glass foam. Several works published in the literature (Axinte et al., 2019; Paunescu et al., 2021; Paunescu et al., 2020) showed the possibility to obtain specific energy consumptions below 1 kWh/kg (up to 0.8 kWh/kg). According to Kharissova et al., 2010, the use of an industrial microwave equipment would increase the process energy efficiency and would reduce the energy consumption by up to 25 %, but the mentioned research is still in an experimental stage.

In recent years, several concrete foaming methods were tested, all performed at room temperature. Synthetic and protein foaming agents were used to obtain lightweight porous concrete, with thermal insulation properties, fireproof and acoustic proof. One of the foaming methods experienced by Kaneshira et al., 2013, is based on the use of gaseous hydrogen as a foaming agent resulting from the corrosion process of aluminum powder in aqueous calcium hydroxide solution (Ca(OH)2). The reactions with water of aluminum oxide (Al2O3), existing in an extremely thin layer on the walls of aluminum particles, produce aluminum hydroxide which reacts with Ca2+ ions in the aqueous solution and forms katoite (Ca3Al2 (OH)12) and hydrogen occur at room temperature. The sludge formed by mixing the powder raw material with water constitutes the adequate medium for capturing the gaseous hydrogen and forming gas bubbles, which by solidification create the foam porous structure. Considering the possibility of foaming the material without the need for sintering at high temperature, the solution applied to the production of porous concrete is also interesting in the case of glass foam manufacturing. A small-scale experiment applying the method of foaming glass waste at room temperature with aluminum powder in aqueous medium of Ca (OH)2 has already been performed by Zeren et al., 2017, promising results being presented at an international conference in Italy.

The current work aimed to test the effect of applying the cold foaming method of glass waste with aluminum powder in aqueous solution of Ca(OH)2 and to compare in terms of quality the characteristics of the foam products obtained by this technique with those of glass foams conventionally produced.

Methods

Generally, the method of turning a powder glass into a foam involves the addition of a suitable foaming agent, which releases a gas or gaseous compound by decomposition or a chemical reaction at temperatures above the value of the softening temperature of the glass, which for a commercial glass (soda-lime glass) is between 800-1000 ºC (Scarinci et al., 2005). The foaming agents currently used in sintering/foaming processes are black carbon, coal, glycerol, calcium carbonate, sodium carbonate, dolomite, calcium sulfate, silicon carbide, silicon nitride, aluminum nitride.

The method of cold foaming the glass with aluminum powder in aqueous solution of Ca(OH)2 is completely different from the method of foaming above the softening temperature of the glass. The aqueous solution of Ca(OH)2 has the role of removing the thin layer of Al2O3 from the surface of aluminum particles and initiating the hydration reaction of aluminum. Carboxymethyl cellulose was added into the starting powder mixture as a binder and foam stabilizer (Ergun et al., 2016). The powder mixture of the solid materials was loaded and manually mixed with a metal rod in a cylindrical stainless steel vessel with an inside diameter of 90 mm and a height of 220 mm. The distilled water was then slowly poured into the vessel, the material being continuously mixed with a rod metal provided with an electrically operated metal propeller. The device used in the experiment is shown in Figure 1. The mixing was performed until the foaming of the material was completed.

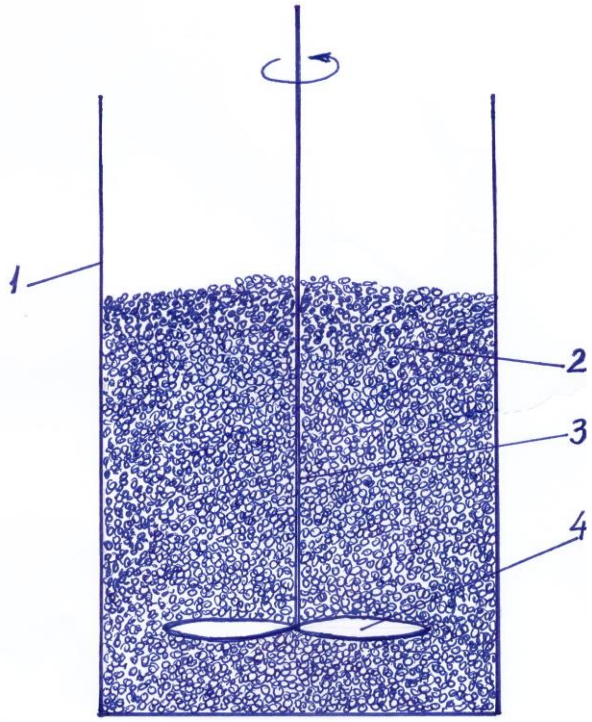

Figure 1. Device for cold foaming the glass-based powder mixture

1 – cylindrical stainless steel vessel; 2 – powder mixture; 3 – metal rod; 4 – metal propeller.

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According to Kaneshira et al., 2003, the chemical reactions that take place at the separation zone between the outer surface of aluminum particles and the aqueous solution of Ca(OH)2 with the successive formation of boehmite (AlOOH), bayerite (Al(OH)3) and aluminum tetrahydroxide (Al(OH)4) and the final release of katoite (Ca3Al2(OH)12) and gaseous aluminum are:

$$\mathrm{Al2O3 + H2O = 2AlOOH} \qquad (1)$$

$$\mathrm{AlOOH + H2O = Al(OH)3} \qquad (2)$$

$$\mathrm{Al(OH)3 + OH- = Al(OH)4} \qquad (3)$$

$$\mathrm{3Ca2+ + 2Al(OH)4 + 3H2O = Ca3Al2(OH)12 + 6H2} \quad (4)$$

To facilitate the formation of hydrogen bubbles and the development of the hydration reaction, the glass-based sludge must be continuously stirred. Thus, the possible solidification of the sludge is prevented. When the material begins the expansion process, the stirring is stopped, its initial volume further increasing until it stops.

The main raw material used in the experiments was a commercial post-consumer packaging bottle (soda-lime glass type) consisting of a mixture of colorless glass and green glass, in equal weight proportions (50/50). The chemical composition (Dragoescu et al., 2018) of the two glass assortments is presented in Table 1.

Table 1. Chemical composition of the glass assortments

Glass type	**Chemical composition, wt. %**							
	SiO_2	Al_2O_3	CaO	MgO	Na_2O	K_2O	Cr_2O_3	Other oxides
Colorless	71.7	1.9	12.0	1.0	13.3	-	0.05	0.05
Green	71.8	1.9	11.8	1.2	13.1	0.1	0.09	0.01

The glass waste processing operations before using in experiments performed in Bilmetal Industries Company in Popesti Leordeni-Ilfov (Romania) consisted of color selection, breaking, grinding in a ball mill and sieving at a granulation of less than 100 μm.

The aluminum powder (grain size below 15 μm) was obtained on an experimental installation at Daily Sourcing & Research SRL Bucharest (Romania) using aluminum waste melt in the microwave field and sprayed with high speed nitrogen jets, according to the own innovative solution (Innovative, 2019).

The Ca(OH)2 powder was purchased from the market at a purity of over 95%. Its role in aqueous solution (approximate concentration 1:6) is to contribute to the release of gaseous hydrogen after dissociation into Ca2+ and OH- and participation in the reactions mentioned above.

Carboxymethyl cellulose (CMC) as a fine powder was also purchased from the market being used as a binder and foam stabilizer.

Usual techniques have been applied in the process of characterizing experimentally manufactured glass foam samples. The gravimetric method (Manual, 1999) was used to measure the apparent density and the comparison method between the apparent density and the density in compact state (true density) of the same material (Anovitz & Cole, 2005) was used to determine the porosity. The use of a TA.XTplus Texture Analyzer allowed to measure the compressive strength and for determining the thermal conductivity the guarded-comparative-longitudinal heat flow (ASTM E1225-04) was applied. The water immersion method (ASTM D570) was used to determine the water absorption of the samples. Using an ASONA 100X Zoom Smartphone Digital Microscope the glass foam microstructures were examined.

The adoption of the variation limits of the weight proportion of the material components took into account, in principle, the experimental results presented in the literature and preliminary own tests. Thus, the aluminum/glass waste ratio was between 0.023-0.078, the Ca(OH)2/aluminum ratio varied between 0.37-0.54, the carboxymethyl cellulose/glass waste ratio was in the range 0.045-0.125 and the distilled water/glass waste ratio was between 0.67-1.00.

Four experimental variants were adopted for the cold manufacture of the glass foam shown in Table 2. The weight ratio of the glass waste varied from 44.8 to 57.2 wt. % and the proportion of aluminum powder was between 1.3-3.5 wt. %, the total amount of dry materials being kept constant at 260 g in all tested variants.

Table 2. Composition of the experimental variants

Variant	**Glass waste wt. %**	**Aluminum powder wt. %**	**$Ca(OH)_2$ powder wt. %**	**Carboxymethyl cellulose wt. %**	**Distilled water wt. %**
1	44.8	3.5	1.3	5.6	44.8
2	50.5	2.8	1.1	4.6	42.0
3	53.6	2.0	0.8	3.5	40.1
4	57.2	1.3	0.7	2.6	38.2

Results and Discussion

The main functional parameters of the cold manufacturing process of glass foam are presented in Table 3.

Table 3. Main functional parameters of the cold manufacturing process

Variant	**Dry/wet raw material amount g**	**Process temperature °C**	**Process duration min**	**Index of volume growth**	**Glass foam amount g**
1	260/376.5	25	16	3.7	256
2	260/369.2	25	17	3.0	257
3	260/364.3	25	17	2.8	257
4	260/359.3	25	18	2.3	258

According to the data in Table 3, the foaming process of glass waste took place at room temperature (25 °C). The foaming process duration was in the range 16-18 min. Using the maximum proportion of aluminium powder (3.5 wt. %) in Ca(OH)2 aqueous solution corresponding to variant 1, the increase by expansion of the material volume was 3.7 times. Lower proportions of aluminium powder (up to 1.3 wt. %) correspond to material expansions more than 2.3 times. Compared to the used amount of dry raw material (260 g), the glass foam amounts represented between 98.5-99.2 %.

Using the methods for determining the physical, thermal, mechanical and morphological characteristics specified above, these features corresponding to each glass foam sample were identified, their values being indicated in Table 4.

Table 4. The physical, thermal, mechanical and morphological characteristics of the glass foam samples

Variant	**Apparent density $g \cdot cm^{-3}$**	**Porosity %**	**Thermal conductivity W/m·K**	**Compressive strength MPa**	**Water absorption vol. %**	**Pore size mm**
1	0.25	88.1	0.059	1.20	2.5	1.9-3.8
2	0.29	86.2	0.067	1.25	2.6	2.0-2.4

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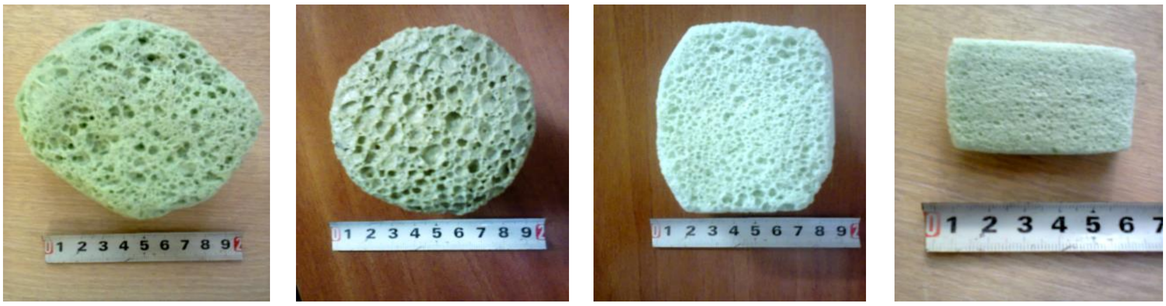
3	0.31	85.2	0.070	1.32	2.4	1.0-1.9
4	0.34	83.8	0.079	1.37	2.6	0.7-1.0

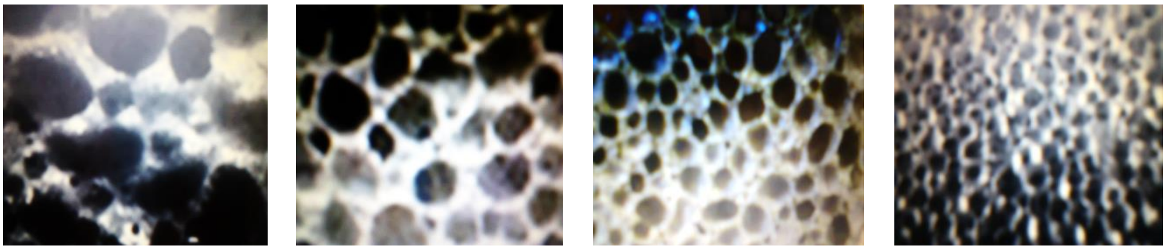
Analyzing the data in Table 4, it can be concluded that the cold manufactured glass foams cold manufactured by the technique described above correspond in terms of quality to products made by conventional hot methods commonly used as thermal insulation materials for inner walls of buildings. The characteristics of the experimental foams correspond to the requirements of the application field mentioned above, i.e. low apparent density (0.25-0.34 g·cm-3), high porosity (83.8-88.1 %), low thermal conductivity (0.059-0.079 W/m·K) which determines the thermal insulation property of the material, compressive strength at an acceptable level (1.20-1.37 MPa) and low water absorption (2.4-2.6 vol. %).

Overall images of the glass foam samples are presented in Figure 1A and cross section pictures of the same samples are shown in Figure 1B. In order to obtain complete information about the glass foam samples, it was necessary to examine their microstructural configuration. Figure 1C shows pictures of the microstructure of the four samples.

A. Overall image of the glass foam samples



B. Cross section of the glass foam samples



C. Microstructural configuration of the glass foam sample______ 4 mm

Sample 1 **Sample 2** **Sample 3** **Sample 4**

Figure 2. Pictures of the glass foam samples
A - overall image; B - cross section; C - microstructural configuration.

The appearance of sample 1 shows an inhomogeneous porosity with rather large pores between 1.9-3.8 mm confirming that the proportion of aluminum as a supplier of the foaming agent (hydrogen) had a too high dosage. In the case of sample 2, the situation is improved, the homogeneity of the pores being visible in Figure 1B. The size of the pores evenly distributed in the section varies in a narrow range (2.0-2.4 mm), but their size can be considered quite large. Sample 3 shows a homogeneous distribution of pores, with dimensions between 1.0-1.9 mm and sample 4 has the most homogeneous distribution of pores, their size being in the range 0.7-1.0 mm. From a microstructural point of view, it can be concluded that samples 3 and 4 are the most suitable as a thermal insulation material. Considering also the physical, thermal and mechanical characteristics from Table 3, sample 3 having the apparent density of 0.31 g·cm-3, the thermal conductivity of 0.070 W/m·K and the compressive strength of 3.2 MPa could be adopted as the optimal glass foam sample.

Therefore, a mixture of 53.6 wt. % colorless and green glass waste, 2.0 wt. % aluminum powder, 0.8 wt. % Ca(OH)2 powder, 3.5 wt. % carboxymethyl cellulose as a binder and foam stabilizer and 40.1 wt. % distilled water is the optimal combination to cold produce a glass foam similar to that manufactured by sintering at high temperature. The main advantage of this process is that significant energy consumption is saved.

The specific energy consumption of melting and overheating at 720 ºC of aluminum in order to spray it in the form of a fine powder must be taken into account. According to Malpohl & Hillen, 2010, the energy consumption for this operation in a natural gas-fired crucible furnace is between 1.28-1.48 kWh/kg, but it must be taken into account that only approx. 2 % of the amount of aluminum participates to the manufacture of glass foam. So, the energy consumption related to the kilogram of glass foam is only 0.026-0.030 kWh/kg, i.e. negligible compared to the consumption of about.0.8 kWh/kg for the conventional hot manufacture of glass foam from glass waste.

It should be mentioned that the current market price of aluminum is 2380 USD/ton (Trading, 2021), obviously higher compared to the main foaming agents used in the conventional manufacture of glass foam: calcium carbonate (between 230-280 USD/ton) and silicon carbide (up to 1200 USD/ton). But the originality of the manufacturing process designed by the authors is the use of recycled aluminum waste.

Conclusion

The aim of the research was the production of glass foams by an innovative technique with a remarkable economical and energy effectiveness compared to conventional industrially applied techniques. A recently tested method of manufacturing porous concrete at room temperature has been taken up for testing in the foaming process of glass waste. The method consists in the use of aluminum powder in aqueous calcium hydroxide solution which releases gaseous hydrogen in a viscous glass waste-based sludge obtained by mixing solids with distilled water in a vessel. The formed gas bubbles are trapped in the sludge mass and by solidification generate a porous structure similar to glass foam. The authors' contribution to the originality of the research was the use of recycled aluminum waste as well as the original technique of melting and overheating the waste by microwave heating applied during the raw material processing. The temperature at which the manufacturing process took place was 25 ºC and the duration of the process was only 16-18 min. The additional energy consumption for melting and overheating the aluminum waste was 0.026-0.030 kWh/kg, insignificant compared to approx. 0.8 kWh/kg required for the manufacture of glass foam by conventional methods at high temperature. The physical, thermal, mechanical and morphological characteristics of the optimal experimental sample were the apparent density of 0.31 g·cm-3, the thermal conductivity of 0.070 W/m·K and the compressive strength of 1.32 MPa similar in terms of quality with those of glass foams conventional manufactured usable as thermal insulation materials for buildings.

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