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NONCONVENTIONAL MANUFACTURE OF CELLULAR GLASS FROM RECYCLED POST-CONSUMER BOTTLE AND BEECH LEAVES AS A VEGETABLE EXPANDING AGENT

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ABSTRACT: The paper exposes testing results corresponding to the process of manufacture cellular glass from consumed drink glass packaging, a vegetable expanding agent (beech leaves) used for the world premiere, and borax as a flux agent. The process of sintering/expanding the powder material mixture was performed by microwave irradiation at 825-834 ºC, as a process improved by the authors of this paper and applied in previous experiments, unlike the conventional procedures used in processes of the same type. The nonconventional heating has allowed a higher energy efficiency of the process and reduced energy consumption (0.78 kWh/kg). The optimal version of the manufacturing process with 30 % beech leaves, 65 % consumed drink glass packaging, and 5 % borax led to obtaining a cellular glass with features adequate for its use as heat insulating material in civil engineering having density of 0.60 g/cm^3, heat conductivity of 0.124 W/mK, compression resistance of 5.0 MPa, absorbing the water of 1.5 vol. %, and cells dimension between 0.7-1.0 mm.

KEYWORDS: cellular glass, microwave warming, consumed drink glass packaging, beech leaf, expanding agent.

1. INTRODUCTION

The recent energy crisis caused by the Russian-Ukrainian conflict with a serious economic impact primarily on European countries has exacerbated existing problems on climate change due to global warming caused by greenhouse gas pollution [1]. Substituting the polluting gas generation sources with "clean" energy sources is the best solution in this case. On the other hand, reducing the share of high energy consuming technologies by using recycled waste and biomass as raw materials and/or additives for manufacturing similar products in terms of quality is an appropriate process, whose application is ongoing worldwide [2].

The current paper is focused on producing the cellular glass from recycled post-consumer bottle. This porous material with excellent physical, thermal and mechanical properties [3] is a cheaper and more economical alternative solution for thermal insulation building materials based on polystyrene existing on the market. The best known method of foaming bottle is the incorporation into its powder mass of an expanding agent, which at high temperature releases a gas through a reaction (usually decay or oxidizing) forming bubbles in the thermally softened mass of the glass [3]. Expanding agents such as carbonaceous products (black carbon, glycerol, graphite, silicon carbide) and metal carbonates (calcium carbonate and less often sodium carbonate) are most often used in industrial cellular glass manufacturing processes. Experimentally on a small scale, several types of agents were tested such as: organic material containing carbon (sugar, starch, hydrocarbons, organic waste) [3], clam shell [4], egg shell [5], organic residues [6], water glass [7], propyl gallate [8], vegetable products (banana leaves, yerba mate, oak leaves) [9-11].

The industrial and small-scale experimental manufacturing methods of cellular glass mentioned above are conventional based on electrical heating or fossil fuels burning heating. In the last 4-5 years, the Romanian company Daily Sourcing & Research SRL has successfully initiated a plan for experimental testing of cellular glass manufacturing by an own original method of nonconventional microwave warming in the joint experimental base of the companies Daily Sourcing & Research SRL and Cosfel Actual SRL. Although discovered in the late 19th century and applied in the 1930s in transmissions and radars, electromagnetic waves were not used in industrial heating processes of solids until the late 20th century and only in moisture removal and warming at low temperature processes. According to [12], microwave warming is a rapid, "clean", and cost effective process due to its special peculiarities. In recent decades, it has been experimentally discovered that various material types (ceramic, organic, polymeric, glass, metallic, etc.) are adequate for effective microwave warming, however the industrial application is delayed and is still in different testing stages.

Given the economic and ecological conditions at European and global level and the global trend towards finding technologies for the manufacture of materials required for industry, construction, transportation, and daily life based on recycled waste and biomass raw materials, the objective of the present paper was to achieve a cellular glass as a heat insulating material for construction sector from consumed drink glass packaging and beech leaves as a vegetable expanding agent. The use of this biomass is the main originality of this research, although the predominantly direct method of microwave warming already applied in previous own experiments [13] also has a character of originality.

Several species of beech trees are developed in large geographical areas with temperate climates in Europe, Asia and North America. Beech is valued as a fire and ornamental wood, but the information offered by the literature on the use of beech leaves in other applications is completely missing. According to [14], the chemical composition of beech leaves includes 95.2 % carbon, 2.1 % hydrogen, 1.4 % calcium, 0.8 % manganese, and the organic constituents are: lignin 37.2 %, cellulose 32.8 %, and hemicellulose 20.9 %, having the features of lignocellulose as an organic matter. The measurements were performed on leaves litter from Canadian beech forests in the initial phase of leaf deposition on the ground. By comparison with the chemical composition and organic constituents of the oak leaves previously tested as a foaming agent [11] there is a relative similarity between the two leaf types, indicating the possibility of successful use of beech leaves in the foaming process of residual glass.

2. MATERIALS AND METHODS

Analysis of beech leaves with the Perkin-Elmer CHN 2400 analyzer revealed a high proportion of volatile solids (over 70 %), which indicates the existence of lignocellulose and carbon fractions. At temperatures in the range of 80-140 ºC volatiles are broken down into gases (carbon monoxide, carbon dioxide, light hydrocarbons) and tar. Heated to 600 ºC, the tar decomposes into elementary components (carbon, hydrogen, oxygen, nitrogen) [15]. The gases resulting from these reactions contribute to the expansion process of the processed residual glass. Beech leaves as an expanding agent were directly collected from the tree, avoiding degraded leaves. They were then finely chopped and ground in an electrically operated laboratory device. After grinding, the leaves were sieved and selected particle sizes below 100 μm.

The glass-based basic material of the producing process of cellular glass was recycled consumed drink glass packaging differenced by color (colorless, green and brown), implicitly, by small variations of the oxide composition showed in Table 1.

Table 1. Composition of the glass types

Composition	**Glass type (wt. %)**		
	Colorless	Green	Brown
SiO_2	71.7	71.8	71.1
Al_2O_3	1.9	1.9	2.0
CaO	12.0	11.8	12.1
Fe_2O_3	-	-	0.2
MgO	1.0	1.2	1.1
Na_2O	13.3	13.1	13.3
K_2O	-	0.1	0.1
Cr_2O_3	0.05	0.09	-
SO_3	-	-	0.05

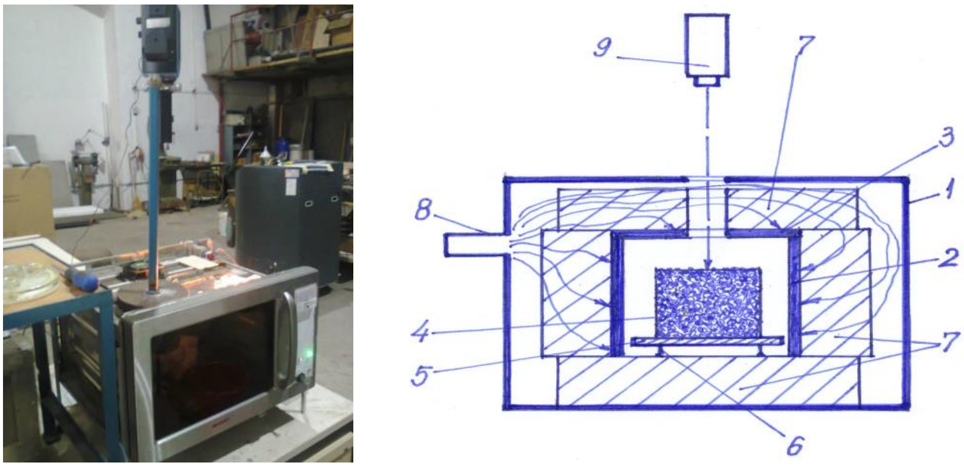
The recycled consumed drink glass packaging was washed, selected by color in equal proportions, broken, shredded in technological equipment for grinding, and granulometrically separated by sieving. The grain size of waste after these processing operations was below 100 μm.

Sodium borate (known as borax) was added to the starting mixture as a flux agent due to its important Na_2O content (30.8 %), considered the best flux material, especially for the glass industry [16]. Also, due to the boron content (about 11 %), borax contributes to a significant increase of the mechanical strength of foamed material [17]. Commercially procured at a grain dimension under 400 μm, it has been necessary to grind it in the electrically operated laboratory device also used in the case of beech leaves. The optimal grain dimension for its use in experiment was under 130 μm.

The experimental microwave plant (Figure 1) mainly consists of 800 W-microwave oven equipped with a single magnetron (microwave generator) of the domestic type. The major difference between the experimental oven and the usual one is the constructive adaptation for warming possibility at much higher thermal level (close to 1200 ºC) and the elimination of the rotation mechanism of heated material due to the use of a significantly higher volume of it compared to the food heating. Under the conditions in which the microwave emission is carried out through a single waveguide, the uniformity of the material warming is not affected due to the high occupancy degree of the useful volume of the oven. The wave propagation in the oven is done completely differently from the distribution of the flue gas after fuel burning through a single burner. All experiments previously performed by microwave warming confirmed the structural homogeneity of material subjected to this heating type.

The direct microwave warming has special peculiarities compared to the conventional warming of solids. According to [18, 19], the initiation of the conversion of microwave power into heat takes place in the middle of material, becoming the hottest area. The heat propagation is done from the inside to the outside, the opposite of the propagation in conventional warming. Also, the direct microwave warming is selective, only the target material (microwave susceptible), being efficiently warmed, unlike the conventional warming process characterized by the initial heating of vault and walls of the oven which then transfers heat to the target material. Previous experiments conducted in 2017 by the authors of the current paper showed that in the case of commercial bottle (soda-lime glass) the direct microwave heating is not adequate due to the too intense contact between material and microwave and excessive high warming rate which causes serious damage in the glass structure at the expanding temperature. This problem was solved by placing a ceramic screen (crucible or tube) made of SiC and Si_3N_4 (materials with high microwave susceptibility) between the wave emission generator and the irradiated material. In this way, the direct heating was slightly tempered and in addition, the ceramic screen partially absorbed the microwave field, then transferring heat by thermal radiation. Thus, a mixed heating system was generated, predominantly direct and partially indirect, which proved to be suitable for highly efficient warming of glass [13].

According to the compositional and operational scheme of the experimental plant (Figure 1b), the ground and pressed glass-based raw material was placed on a stainless steel plate under a ceramic tube covered with a lid. The tube and lid were protected with heat insulating layers of ceramic fiber blankets. The process temperature control was made with a radiation pyrometer mounted above the oven, which visualized the hot material through holes provided in the upper wall of oven and lid.



a b

Figure 1. Experimental microwave plant
a – general image of the experimental plant; b – compositional and operational scheme:
1 – 800 W-microwave oven; 2 – SiC + Si3N4-ceramic tube; 3 – ceramic lid; 4 – pressed mixture;
5 – stainless steel plate; 6 – metal support; 7 – heat insulating; 8 – microwave field waveguide; 9 – pyrometer.

The experimental process of making the cellular glass was performed in four experimental versions containing recycled consumed drink glass packaging (between 45-75 wt. %) as basic material, borax (5 wt. %) as a flux agent, beech leaf (between 20-50 wt. %) as an expanding agent, and water addition (12 wt. %) as a binder. The composition of the four versions is shown in Table 2.

Table 2. Composition of the starting mixture

Composition	**Version (wt. %)**			
	1	2	3	4
Consumed drink glass packaging	75	65	55	45
Borax	5	5	5	5
Beech leaf	20	30	40	50
Water				

addition	12	12	12	12

Determining the characteristics of the cellular glass samples was performed using common methods of analysis. The apparent density was measured by the gravimetric method [20] and the porosity was calculated by the method of comparing the true and apparent density [21]. The compressive strength was determined using a TA.XTplus Texture Analyzer and the thermal conductivity by the guarded-comparative-longitudinal heat flow method (ASTM E1225-04). The water absorption was measured by the water immersion method (ASTM D570) and the samples microstructure was investigated with a Smartphone Digital Microscope.

3. RESULTS AND DISCUSSION

3.1 Results

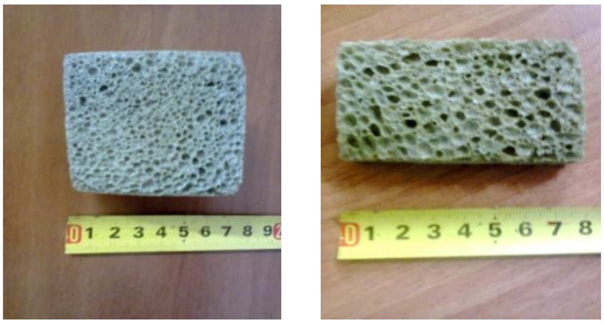
According to Table 3, 480 g of dry material mixture having the composition in Table 2 were successively sintered at temperatures between 825-834 ºC corresponding to each of the four experimental versions.

Table 3. Functional parameters of the manufacturing process

Parameter	Version 1	Version 2	Version 3	Version 4
Dry raw material amount (g)	480	480	480	480
Process temperature (ºC)	825	828	831	834
Warming time (min)	33	35	37	40
Average rate (ºC/min)				
- warming	24.4	23.1	21.9	20.4
- cooling	5.2	5.2	5.3	5.2
Cellular glass amount (g)	465	466	464	465
Index of volume increasing	1.20	1.40	1.70	2.10
Specific consumption of electricity (kWh/kg)	0.74	0.78	0.83	0.90

The warming rate of the mixture had high value between 20.4-24.4 ºC/min significantly above the rates level developed in the process of making cellular glass with oak leaves as an expanding agent (between 15.1-17.8 ºC/min) [11]. The specific energy consumption had economical values between 0.74-0.90 kWh/kg. The material expansion using beech leaves reached higher values (2.10) compared to those foamed with oak leaves (1.75).

The appearance of the cross-sectional cellular glass specimens obtained by foaming the consumed glass packading with borax and beech leaves is shown in Figure 2.



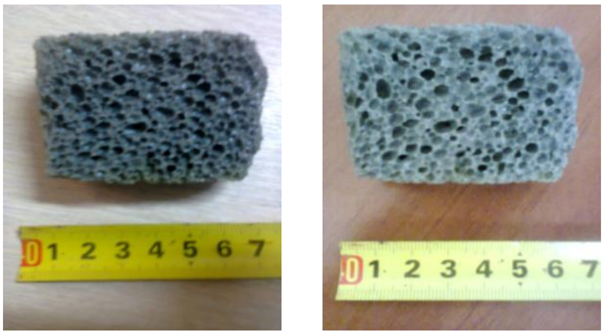


Figure 2: Cross section of the cellular glass specimens
a – version 1; b – version 2; c – version 3; d – version 4.

Generally, the macrostructure of cellular glass specimens is homogeneous enough to ensure materials with heat insulating properties required for building applications. The macrostructural fineness in the case of version 1, the best homogeneity of the pore distribution in section in the case of version 2, and then the tendency towards slightly coarse structures in the case of specimens 3 and especially 4 can be observed.

The main physical, thermal, mechanical and morphological features of the cellular glass specimens are presented in Table 4.

Table 4. Features of the cellular glass specimens

Feature	Version			
	1	2	3	4
Apparent density (g/cm^3)	0.92	0.60	0.41	0.31

Porosity (%)	56.2	71.4	80.5	85.2
Heat conductivity (W/mK)	0.210	0.124	0.089	0.075
Compression strength (MPa)	8.3	5.0	3.1	1.8
Water absorbing (vol. %)	1.5	1.5	1.3	1.0
Cell size (mm)	0.5-0.9	0.7-1.0	0.8-1.9	1.0-2.0

In the four tested versions, the weight proportions of consumed glass packaging and beech leaves were variables, because the borax ratio was kept constant. The beech leave proportion as an expanding agent was successively 20, 30, 40, and 50 %, while the glass proportion decreased from 75 to 45 %. According to the data in Table 4, it was observed that a low beech leaves/glass ratio (20/75) favors obtaining a dense material (0.92 g/cm^3) with low cells (0.5-0.9 mm) and very high compression strength (8.3 MPa), while a high ratio (50/45) leads to obtaining a porous product (0.31 g/cm^3) with coarse structure (cell dimension between 1-2 mm) and relatively low compression strength (1.8 MPa). The optimal version was considered version 2 with moderate heat insulating properties (apparent density of 0.60 g/cm^3, heat conductivity of 0.124 W/mK, porosity of 71.4 %), high compression resistance (5.0 MPa), homogeneous structure (cell size between 0.7-1.0 mm), and a low specific energy consumption (0.78 kWh/kg) for producing this material.

The microstructural configuration of the cellular glass specimens is shown in Figure 3, which confirms the uniform distribution of cells in versions 1 and 2, their relatively coarse distribution in versions 3 and 4, as well as the range of cell size corresponding to the four specimens.

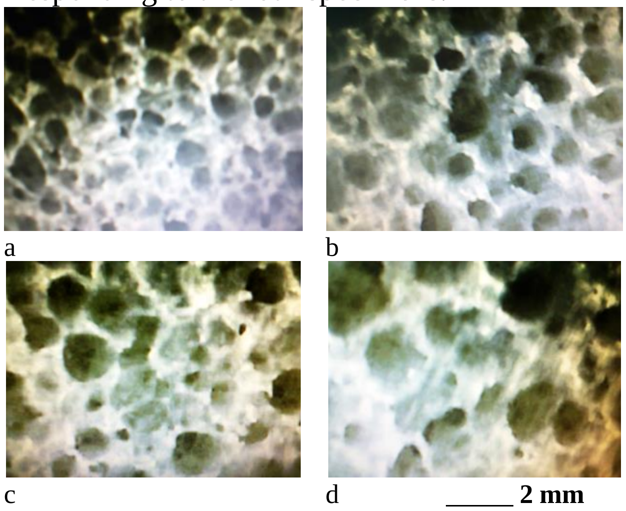


Figure 3: Microstructural images of the cell glass specimens a – version 1; b – version 2; c – version 3; d – version 4.

3.2 Discussion

The experimental results showed the ability of beech leaves as an expanding vegetable agent to foam the soda-lime residual glass. The sintering/expanding temperature between 825-834 ºC is in the range of similar process temperatures with carbon (750-800 ºC) and calcium carbonate (800-900 ºC) and are lower than the expanding temperatures with silicon carbide (over 950 ºC). The use of the nonconventional microwave warming technique allowed reaching significantly higher warming rates (over 20 ºC/min) compared to recommended and frequently used in conventional processes (around 10 ºC/min), without affecting the microstructural homogeneity of the foamed product. The physical, thermal and mechanical features of cellular glasses experimentally making with beech leaves and consumed glass packaging falls within the required limits for heat insulating materials for building. In the experiment presented in the paper, the addition of borax as a flux agent often used especially in the case of calcium carbonate foaming, contributed to the increase of the compression resistance of cellular glass up to 8.3 MPa (due to the high enough boron content). According to the literature, the use of some vegetable foaming agents (banana leaves, oak leaves or yerba mate) contributed also to obtaining high values of compression resistance of maximum 3.5; 3.4 or even 15 MPa (in case of using yerba mate).

4. CONCLUSION

A new vegetable expanding agent (beech leaf) was tested in the process of making the cellular glass from recycled soda-lime residual glass using the nonconventional predominantly direct microwave warming method and this was the aim of the paper. Only the oak leaf was used also as an expanding agent embedded in the residual glass powder warmed by the mentioned nonconventional method, unlike banana leaf and yerba mate used in conventional making processes of cellular glass. The paper is original both in terms of the type of vegetable agent tested for the first time in the world and the used warming method, which is typical of the team of current paper authors, although the method has already been used in several previous experiments of this team. The functional parameters of the sintering/expanding process had the temperature range between 825-834 ºC, the warming rate being much higher (over 20 ºC/min) compared to conventional processes, without affecting the macro- and microstructural homogeneity of foamed products. The characteristics of the optimal cellular glass (version 2) were: apparent density of 0.60 g/cm^3, heat conductivity of 0.124 W/mK, porosity of

71.4 %, compression strength of 5.0 MPa, water absorbing of 1.5 vol. %, and cell size between 0.7-1.0 mm. The specific energy consumption (0.78 kWh/kg) for manufacturing this material was very low. The cellular glass made of recycled residual glass, beech leaves, and borax is a cheaper and more economical alternative solution for heat insulating building materials.

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