THERMAL INSULATING MATERIAL WITH HIGH MECHANICAL STRENGTH MADE FROM CLAY BRICK WASTE AND COAL ASH USING THE MICROWAVE ENERGY

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Abstract: The paper presents the results of experimental research for manufacture a high mechanical strength thermal insulating material using the microwave energy. Clay brick waste (75 - 83 mass %) and coal ash (15 - 23 mass %) as raw material and silicon carbide (2%) as a foaming agent have been used as a powder mixture. The porous product obtained by a sintering/ foaming process at 1115 - 1145 ºC had relatively low density and thermal conductivity (0.50 - 0.68 g/cm^3 and 0.078 - 0.095 W/mK, respectively) and high compressive strength (up to 7.5 MPa). This remarkable combination of some physical and mechanical characteristics of the insulating material allows its use in applications involving mechanical stress resistance.

Keywords: aluminosilicate waste, clay brick waste, coal ash, glass-ceramic foam, microwave, high mechanical strength

1. INTRODUCTION

Clay brick waste from demolition and rehabilitation of buildings represents a very large amount of raw material and a consistent annual generation rate available for material recycling processes. On the other hand, coal ash (as fly or bottom ash) results in large quantities from combustion processes specific to the thermoelectric power stations. The mixing of the two finely ground aluminosilicate wastes, the addition of a foaming agent and the thermal treatment at high temperature (over 1100 °C) allow to obtain a porous product with high mechanical strength, usable as a building insulating material, in areas involving simultaneous association of low density and mechanical strength (pavements, road construction, outer panels, drainage, sports grounds, foundations, aggregate for lightweight concrete, etc.).

The use in the sintering/foaming process of aluminosilicate wastes such as coal ash, metallurgical slag, mud from zinc hydrometallurgy, fly ash and dust from incinerator waste, sludge, etc. favors the formation of a rigid raw material matrix at the softening temperature, contributing to the increase of the structure resistance of the foam after cooling. The product is a glass-ceramic foam, characterized by the existence at least of a crystalline phase (obtained by a controlled crystallization of the silicate) and one amorphous [1, 2].

The literature does not directly provide information about the simultaneous influence of clay brick waste and coal ash on the mechanical and physical properties of the cellular products resulted by sintering/ foaming of some mixtures containing these wastes, although clay as a mineral is frequently associated with coal ash [3]. In

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principle, the clay from brick waste contributes to the significant increasing of the mechanical strength of the products [4-7] and the coal ash maintains a relative low level of the values of apparent density and thermal conductivity [7].

Worldwide, the common heating techniques at the sintering/ foaming temperature of silicate waste, especially, of aluminosilicate waste, such as clay brick waste and coal ash, are conventional (fossil fuel consumption or electrical resistances). Theoretically, the use of microwave energy would not be adequate due to their high content of SiO_2 and Al_2O_3, that are microwave transparent materials and, for this reason, the heating process of these waste is not effective at low temperature (up to 500 ºC) [8-10]. In reality, it was experimentally confirmed by the company Daily Sourcing & Research [11] that the presence in the wastes composition of Fe_2O_3 and Cr_2O_3 compensates this behavioral disadvantage and the materials above mentioned can be fast and effective heated starting from the room temperature [9, 10]. The main advantages of the heating by the microwave irradiation compared to the conventional heating are higher heating rates, heating selectivity, better process control and energy saving. The microwave susceptible material absorbs the microwave energy in its entire volume and transform it into heat. Because the material itself generates heat, the heating process occurs volumetrically and can be very fast [12]. High concentrations of alkali metal oxides (Na_2O, K_2O) in the composition of a material directly microwave heated strongly favor the microwave absorption due to the correlation between the electrical conductivity of the material and its absorbent capacity [10]. In case of sintering/ foaming processes based on commercial glass, the Na_2O mass ratio exceeds 13% and could be the main reason why the microwave irradiation severely affects the macrostructure of these types of waste through the excessively high heating rate starting from the core of the material [11]. In the case of aluminosilicate wastes (clay and coal ash) the proportion of Na_2O and K_2O is significantly lower (1-6% and 4.1 - 4.4%, respectively) and it is believed that this could be the reason why the radiation absorption aggressiveness is diminished below the limit of macrostructural destruction. The heating method adopted in the research presented in the paper is unconventional, using the microwave energy. This technique, which has been used to a very small extent up to ten years ago, especially in the domestic food preparation field and in a few other areas (vulcanization of rubber and manufacture of polymer/ wood composites), has seen a relatively increasing trend in recent years, after it has been found that the microwave energy can be used effectively for other types of materials: ceramics, organics, polymers, metals, glass, sol-gel, composites, etc. [13]. In a recent paper [14], the team of researchers from Daily Sourcing & Research used in the raw material powder mixture old clay brick waste (22-35.2%), coal ash (9%) and green container glass waste (52.8 – 66%), together with silicon carbide (3%) as a foaming agent. The mixture was sintered/ foamed by indirect microwave heating at temperatures between 1000-1060 ºC, with heating rates between 16.8 - 18.5 ºC/min, obtaining a glass-ceramic foam with the maximum compressive strength of 2.63 MPa, the apparent density of 0.70 g/cm^3 and the thermal conductivity of 0.094 W/m·K. The specific energy consumption had values between 2.91- 3.46 kWh/kg.

The research objective whose results are presented below was the use of some waste generated by buildings demolition (clay brick waste) to produce a high mechanical strength insulating material by sintering and foaming at over 1100 ºC. The adopted manufacturing technique is original being used the direct microwave heating. This very advantageous technique is not suitable for any material type or materials mixture. The team of the authors of this paper experimentally found that the clay brick waste can be heated by the direct microwave irradiation without affecting its internal structure. Thus, the foaming process becomes much faster than any used conventional technique and the specific energy consumption can be significantly reduced.

2. METHODS AND MATERIALS

2.1. Methods

In the sintering/foaming processes of the glass-based mixtures, the heating into a 0.8 kW-microwave oven could not be carried out directly due to the destruction of the internal structure of material under the influence of the electromagnetic radiation. Experiments conducted in the company Daily Sourcing & Research led to obtaining high quality foamed products only applying indirect heating methods by the placement of a cylindrical tube or crucible made of silicon carbide (a microwave susceptible material) between the microwave emission source and the material [11, 15, 16]. Unlike the processes above described, the foaming of the powder mixture containing aluminosilicate wastes (clay brick waste and coal ash) was directly made by the contact between the microwave field and the mixture. The pressed material, protected with ceramic fiber mattresses (Figure 1,b) both on the side surface, at the bottom and on the upper surface, to avoid heat loss outside the system, was introduced into the oven (Figure 1.c). The only microwave generator (0.8 kW) has the waveguide placed on one of the sidewalls of the oven. The heating uniformity was achieved by rotating the material around its own vertical axis with a mechanism adapted to the high temperature conditions. The thermal process control was performed with a Pyrovar radiation pyrometer (Figure 1 a point 3) mounted above the oven on a metal support (Figure 1 a point 2), viewing the heated material surface through the hole from the upper metal wall of the oven and that from the upper ceramic fiber layer.

a b c

Fig. 1. Experimental microwave equipment: a-0.8 kW-microwave oven; a point 1 – oven; a point 2-metal support; a point 3-pyrometer; b-the pressed powder mixture coated with ceramic fiber; c-positioning the thermal protected material into the oven.

2.2. Materials

Two aluminosilicates (clay brick waste and coal ash) constituted the raw material used in the experiments. Due to the low amounts of clay brick waste, its processing was performed manually by crushing, followed by the dimensional sorting by sieving below 250 μm. The coal ash purchased from the Paroseni Romanian thermoelectric power station was sieved at dimensions below 100 μm. The adopted foaming agent was silicon carbide bought from the market at the grain size below 40 μm and used as such. The chemical composition of the raw material is shown in Table 1.

Table 1. Chemical composition of clay brick waste and coal ash.

Material	Chemical composition, mass %						
	SiO_2	Al_2O_3	CaO	Fe_2O_3	MgO	Na_2O	K_2O
Clay brick waste	56.4	27.4	1.2	7.2	1.4	1.0	4.4
Coal ash	46.5	23.7	7.9	8.6	3.2	6.0	4.1

The XRD pattern of clay brick waste [17] indicated as crystalline phases: quartz, muscovite, clinochlore, calcite and sanidine. The XRD analysis of coal ash [18] showed the following crystalline phases: quartz, mullite, illite, kaolinite and calcite.

The raw material and the foaming agent were dosed in variable mass ratios, constituting four compositional variants for experiments. A supplementary water addition in the constant mass ratio of 25% (over 100%) completed each of the four batches of materials. Table 2 presents the experimental batches composition.

Table 2. Composition of the experimental batches of materials.

Raw material	Variant			
	1	2	3	4
Clay waste, mass%	75	78	80	83
Coal ash, mass%	23	20	18	15
Silicon carbide, mass%	2	2	2	2
Water addition, mass%	25	25	25	25

To confirm the reproducibility of the samples, the four variants were made in two sets. Also, one of the sets was subjected to the destructive test (by cracking) for determining the compressive strength.

2.3. Characterization of the samples

The glass-ceramic foam samples, made by the sintering and foaming experimental process, were tested in laboratory to identify the physical, mechanical and morphological characteristics. The apparent density was measured by the gravimetric method [19]. The porosity was calculated by the comparison method of the density of the compact material (melted and cooled) and the density of the porous material, experimentally measured [20]. The water absorption of the sample was determined by the method of its water immersion. The thermal conductivity was determined by the guarded-comparative-longitudinal heat flow technique by measuring the thermal flow that passes through the ceramic sample (140 x 140 x 50 mm) placed between two metal plates, one heated and thermally protected and the other cooled. To determine the compressive strength, a device built according to the own design was used, which develops an axial pressing force exerted by the hydraulically operated piston of maximum 20 tf and can measure axial compressive strengths up to 40 MPa. The sample has a cylindrical shape with a diameter of 80 mm and a height of 70 mm. The test measures the value of the compressive strength of the sample before cracking. The identification of the crystalline phases of the glass-ceramic foams was performed according to the standard EN 13925 – 2: 2003 with an X-ray diffractometer Bruker AXS Advance with CuK α radiation. The microstructure investigation of the samples was performed with a Smartphone digital microscope.

3. RESULTS AND DISCUSSION

The main functional parameters of the foaming process, containing the amounts of the dry raw material and the cellular product, the process temperature and its heating duration, the average heating and cooling rates, the index of volume growth and the specific energy consumption are presented in Table 3.

The temperature of the sintered and foamed powder mixture was measured with the radiation pyrometer. The indication for stopping the microwave heating process due to the sample foaming was the stabilization of the temperature of visualized sample surface and the beginning its decreasing. The duration of the heating process was determined by this moment of stopping the operation of the microwave generator. The average heating rate was calculated as the ratio between the difference between the final and the initial temperature and the duration of the heating process. The average cooling rate was determined as the ratio of the temperature loss between the maximum and minimum values and the time required for cooling. Usually the minimum temperature value was 40 - 50 ºC. The index of volume growth represented the percentage increase of the sample volume by foaming compared to the initial volume of the raw sample. The electricity consumption was counted, and the mass of the cold foamed product was weighed to determine the specific energy consumption.

Table 3. Parameters of the foaming process.

Var.	Dry raw material/ foamed product amount, g	Foaming temperature, ºC	Heating time, min	Average rate, ºC/min		Index of volume growth	Specific energy consumption, kWh/kg
				Heating	Cooling		
1	580/561	1115	31	35.3	6.1	1.40	0.74
2	580/559	1120	32	34.4	5.9	1.30	0.76
3	580/563	1130	34	32.6	5.9	1.25	0.81
4	580/565	1145	36	31.3	6.3	1.20	0.85

As a feature of microwave direct heating processes, which are not suitable for glass (as mentioned in chapter 2.1), the average heating rate of the material has reached considerably higher values (up to 35.3 ºC/min) compared to the rates usually used in the glass waste foaming (between 12 - 24 ºC/min), without its internal structure being destroyed. By increasing the mass ratio of clay waste from 75 to 83%, the temperature of the sintering/ foaming process increased in the range 1115 - 1145 ºC and implicitly, the duration of the process (from 31 to 36 minutes). As a consequence, the heating rate decreased slightly from 35.3 to 31.3 ºC/ min and the specific energy consumption increased from 0.74 to 0.85 kWh/kg. It should be noted that, by comparison with own experimental results obtained in indirect heating processes using the same type of unconventional energy [21], the duration of the microwave direct heating process was drastically reduced, almost half. Unlike the manufacturing processes of glass foam from glass waste, characterized by high values of volume growth index of over 2.5 [15, 16], in the case of using clay waste and coal ash, this parameter had significantly lower values (between 1.2 - 1.4), the lowest value corresponding to the variant with the highest clay ratio.

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

Table 4. Physical, mechanical and morphological features of the samples.

Variant	Apparent density, g/cm^3	Porosity, %	Thermal conductivity, W/mK	Compressive strength, MPa	Water absorption, %	Pore size, mm
1	0.50	59.5	0.078	3.8	12.3	1.0 – 5.0
2	0.59	63.2	0.084	5.1	11.9	0.9 – 2.5
3	0.61	65.9	0.088	6.3	12.7	0.8 – 2.0
4	0.68	68.6	0.095	7.5	12.8	0.7 – 2.0

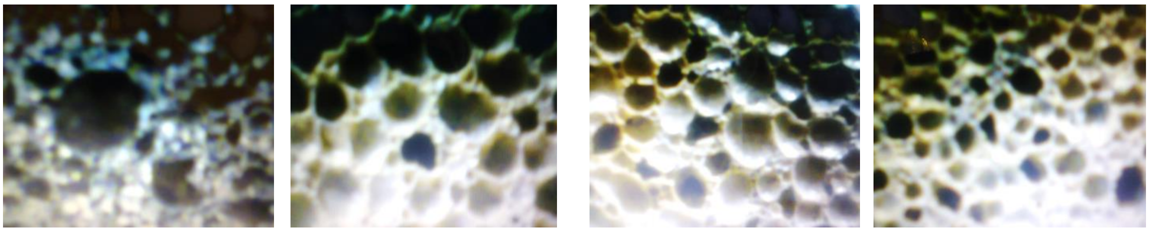
Considering that the cellular material produced from aluminosilicate waste (clay brick and coal ash) was intended for use in construction as a thermal insulation material for applications involving mechanical stress resistance, the main features of particular interest were compressive strength, apparent density and thermal conductivity. The compressive strength has reached high values for a material with thermal insulation characteristics (between 3.8-7.5 MPa) compared to the values of similar products manufactured industrially from glass waste (up to 6 MPa) [22, 23]. The apparent density (maximum 0.68 g/cm^3) and the thermal conductivity (maximum 0.095 W/mK) corresponding to the highest value of compressive strength are relative low, being acceptable for the application domain. This combination between the physical and mechanical features of the samples is appropriate for the applications mentioned above. Considering that clay is a highly water-absorbing material, the range between 11.9-12.8% of the water absorption of the glass-ceramic foam obtained experimentally is considered acceptable. By comparison, lightweight aggregates from masonry rubble presented in the literature have the water absorption between 6-16%, while expanded clay, a lightweight aggregate currently used in the world, has these limits far higher (14-26%) [7]. The pore size range decreases from sample 1, containing the minimum mass ratio (75%) of clay waste (1.0-5.0 mm) up to sample 4, containing the maximum mass ratio (83%) of clay waste (0.7-2.0 mm). Generally, the pores are uniformly distributed, forming a homogeneous porous structure. Images of the cross section of the four glass-ceramic foam samples are shown in Figure 2.

a b c d

Fig. 2. Pictures of the cross section of the glass-ceramic foam samples: a-sample 1; b-sample 2; c-sample 3; d-sample 4.

The cross sections of the porous samples (Figure 2) indicate a significant homogenization of pores repartition and diminishing the pore size between the sample 1 and the other three, due to the reduction of the weight ratio of coal ash and simultaneous, the increase of the ratio of clay waste.

The microstructural analysis of the porous samples was performed with a Smartphone Digital Microscope. The pores sizes in the cross section could be determined (Table 4) both with this device, but also at the true dimension of the samples by direct measurement. Microstructural images corresponding to the four samples are shown in Figure 3.



a b c d

Fig. 3. Microstructural configuration of the samples: a-sample 1; b-sample 2; c-sample 3; d-sample 4.

The XRD analysis allowed the identification of the crystalline phases of the samples after the thermal treatment. The main crystalline phases were quartz and, in smaller proportions, mullite, illite, kaolinite and calcite. Figure 4 shows a representative diagram containing the XRD analysis of sample 4, with the highest compressive strength value, heat treated at 1145 ºC.

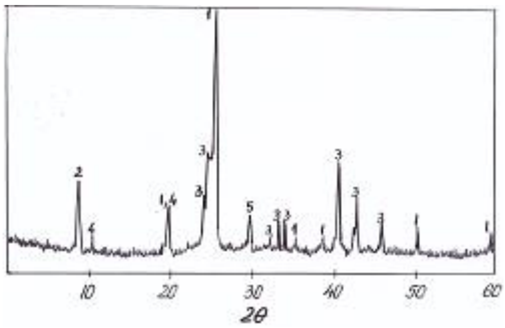


Fig. 4. XRD analysis for the sample 4 heated at 1145 ºC: 1–quartz; 2–illite; 3–mullite; 4–kaolinite; 5–calcite.

A special analysis should be made regarding the energy efficiency of the manufacturing process of glass-ceramic foam with high mechanical strength from aluminosilicate waste by direct microwave heating. Generally, the specific consumption of the industrially made products from glass waste (i.e. foam glass gravel) by conventional heating methods is not provided by the literature. However, an information due to a market study [23] indicates, indirectly, an average consumption of the Misapor consortium, mainly specialized on such porous products with high mechanical strength (up to 6 MPa), of 100 kWh/m^3, i.e. about 0.85 kWh/kg. According to Table 3, the specific energy consumption achieved in the experiments described in the paper had values between 0.74-0.85 kWh/kg. If one considers the major differences between a continuous industrial process and a discontinuous experimental process performed on a very low power equipment (0.8 kW), it can be concluded that the direct microwave heating offers a significantly higher energy efficiency.

4. CONCLUSIONS

A glass-ceramic foam from aluminosilicate wastes (clay brick and coal ash) was made in the company Daily Sourcing & Research by direct microwave heating at high temperatures between 1115-1145 ºC.

The obtained foam had physical, mechanical and morphological features adequate for its use in construction as an insulating material under high mechanical stress conditions as replacer of the materials existing on the market. The highest compressive strength value (7.5 MPa) of the glass-ceramic foam made from 83% clay waste, 15% coal ash as raw material and 2% silicon carbide as a foaming agent was obtained under conditions in which the apparent density and the thermal conductivity had relative low values (0.68 g/cm^3 and 0.095 W/ m K respectively), considered acceptable for the application domain.

The macroscopic appearance of the cross sections of the samples was homogeneous with a uniform distribution of the pores. In the case of the sample with the highest compressive strength, the pore size varied between 0.7 - 2.0 mm.

Due to the direct microwave heating system, the heating rate had very high values between 31.3-35.3 ºC/min and the specific energy consumption were very low (between 0.74- 0.85 kWh/kg), demonstrating the remarkable energy efficiency of the manufacturing process of the glass-ceramic foam from aluminosilicate wastes through this unconventional method. Practically, all the tested variants correspond to the proposed purpose of achieving a material that can be used as an insulator under mechanical stress conditions. Obviously, the sample 4 with the highest compressive strength is recommended for applications that involve a maximum requirement.

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