LIGHTWEIGHT AGGREGATE MADE BY MICROWAVE IRRADIATION

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ABSTRACT

The paper presents experimental results of the manufacture of a lightweight aggregate from a waste (recycled clay brick from the demolition of buildings) and an industrial by-product (coal ash) using the unconventional microwave heating technique. The manufacturing process is based on the direct microwave heating at high temperature (over 1160 °C) of the finely ground mixture of the raw material having incorporated a foaming agent (silicon carbide) with a role in expanding the material. The manufactured aggregate is a porous product with low apparent density and low thermal conductivity (below 0.80 g/cm³ and below 0.148 W/m·K, respectively), high mechanical strength (up to 8 MPa), low water absorption (below 7%), being suitable for the production of lightweight concrete. The specific energy consumption of the thermal process is very low (0.83-0.89 kWh/kg).

Keywords: lightweight aggregate; direct microwave heating; coal ash; clay brick waste; mechanical strength

1. INTRODUCTION

The lightweight aggregates used as raw material in the manufacture of mortars and concretes can come from natural resources (volcanic synthetically rock) or are manufactured by thermal processes at high temperatures from natural waste or industrial by-products (clay, slate, coal ash, sewage sludge, etc.). The basic principle of the manufacture of lightweight aggregates consists in the use of a foaming agent embedded in the mixture of raw material to release a gas through a chemical reaction of oxidation or decomposition in the thermally softened mass of the material. Its adequate viscosity blocks the gas in the form of bubbles and the subsequent cooling of the material facilitates

REZUMAT

Lucrarea prezintă rezultate experimentale ale fabricării unui agregat ușor dintr-un deșeu (cărămidă de argilă reciclată din demolarea clădirilor) si un produs secundar industrial (cenusă de cărbune) utilizând tehnica neconventională a încălzirii cu microunde. Procesul de fabricare se bazează pe încălzirea directă cu microunde la temperatură înaltă (peste 1160 °C) a amestecului fin măcinat al materiei prime având înglobat un agent de spumare (carbură de siliciu) cu rol în expandarea materialului. Agregatul fabricat este un produs poros cu densitatea aparentă și conductivitatea termică reduse (sub 0,80 g/cm³ și, respectiv, sub 0,148 W/m·K), rezistență mecanică mare (până la 8 MPa), absorbtie a apei în limite mici (sub 7%), fiind adecvat pentru producerea betonului ușor. Consumul specific de energie al procesului termic este foarte mic (0,83-0,89 kWh/kg).

Cuvinte cheie: agregat ușor; încălzire directă cu microunde; cenușă de cărbune; deșeu de cărămidă de argilă; rezistență mecanică

the formation of a light porous structure (Moeller et al., 2015).

From the data presented in the literature (Hammer et al., 2000), lightweight aggregate manufacturing techniques are known. The most used are lightweight expanded clay aggregates (LECA), manufactured by heat treatment at about 1200 °C of clay in a rotary kiln. The foaming gas expands the clay by numerous bubbles. The manufacturing technique of LECA has been developed since the first decades of the 20th century in the United States. In Europe, LECA has started to Denmark, Germany, produced in Netherlands, Great Britain and in Central and Eastern European countries. The aggregate is manufactured in the form of pellets and is available in different sizes between 0.1 and 25 mm and apparent densities between 0.25 and 0.51 g/cm³. The LECA aggregates provide lightness, thermal insulation due to the thermal conductivity below 0.097 W/m·K, soundproofing by acoustic resistance, moisture impermeability, fire resistance and frost-thaw resistance. They are also suitable as a material for drainage and filtration.

Some information in the literature (Moeller et al., 2015) refers to the manufacture of lightweight aggregates from recycled masonry rubble. Using different weight ratios of the waste components (concrete, natural aggregate, clay brick, and other mineral components) resulted in mixtures of raw material with a density ranging between 1.95 and 2.42 g/cm³. The oxide composition of the mixtures contained mainly SiO₂ (67.7-76.7%), Al₂O₃ (8.6-12.0%) and CaO (8.2-11.4%) as well as small ratios of Fe₂O₃, MgO, K₂O, Na₂O and SO₃. The raw material was crushed, screened, ground, mixed and fired at 1160-1180 °C in a laboratory rotary kiln. Silicon carbide was used (between 1-3%) as a foaming agent. The bulk density of aggregate as pellet was between 0.7 and 1.0 g/cm³ and the compressive strength had values between 4 and 5 MPa. The water absorption of samples measured by water immersion for 24 hours was 8-15.5%, being significantly lower than that of expanded clay aggregate (14-25%).

The raw material used in the paper (Jacubcová et al., 2011) was also recycled masonry rubble containing mainly clay brick, mortar and plaster as well as other materials, such as lightweight concrete, usual concrete, sand lime brick and aerated concrete. The rubble mixture was crushed and ground in a ball mill at grain size under 100 µm. The prepared powder was mixed with silicon carbide as a foaming agent and water in a high shear mixer, which allowed mixing and granulation at the same time. The raw granules were expanded and stabilized in a rotary kiln. A dosage of 3% silicon carbide and a firing temperature of about 1185 °C led to the maximum expansion effect. The comparison of the lightweight aggregate made from masonry rubble with conventional aggregates of expanded clay did not lead to significant differences.

The paper (Kayali, 2005) presents an innovative technique for manufacturing a lightweight aggregate in which the only solid waste used is fly ash. Sintered pelletized fly ash aggregates have been produced in several countries such as UK, USA, Japan, India and countries of Northern Europe. The product manufactured according to the patent is unique, easy to manufacture and requires low energy. The temperature of the sintering process of the fly ash-based material is between 1090 and 1370 °C. The pelletizing is not used, the sintered material being broken to the required dimensions. The new aggregate type has a bulk density of 0.84-0.86 g/cm³ and the water absorption in 24 hours is around only 3%. According to (Kayali, 2005), the concrete made with this type of aggregate is 21% lighter, has a mechanical strength 15-21% higher and has a shrinkage value at least 30% lower than the normal-weight concrete.

The paper (Gao et al., 2012) presents experimental results obtained manufacturing process of a sintered fly ash aggregate. The mechanical strength of the product was 7.8 MPa and the water absorption was 4.2%. The basic raw material was fly ash and the auxiliary raw materials were clay, a melted sludge, coal and paper-mill waste. The stages of the manufacturing process included: drying, grinding and mixing the raw material, pelletizing, desiccating, pre-sintering and sintering the raw pellets. The expansion of the material took place due to the release of CO₂ following the reaction between C in coal and Fe₂O₃. The pre-sintering temperature was 700 °C for 25 min and the temperature was 1250 °C for 20 min.

A technical solution for the production with very low energy consumption (in the microwave field) of a lightweight aggregate by using the masonry rubble from buildings demolition and rehabilitation is presented in the paper (Paunescu et al., 2019). The masonry rubble (85.6-90.8%) had an average composition including clay brick, concrete and cement mortar in the weight ratio 48/40/12. Coal ash (between 4.0 and 9.0 wt.%) was an additional material. Silicon carbide (between 3.5 and 5.0 wt.%) was selected as foaming

agent and the water addition was kept constant at 18 wt.%. The required sintering temperature had values between 1168 and 1185 °C. Due to the direct microwave heating at very high speeds (between 33.9 and 35.9 °C/min), the process duration was relatively short (32-34 min). The resulting aggregate had the apparent density between 0.75 and 0.98 g/cm³, the porosity between 60.8 and 70.0%, the thermal conductivity in the range 0.123-0.140 W/m·K, the compressive strength between 6.0 and 7.3 MPa, the water absorption between 12.9 and 14.6% and the pore size in the limits 0.8-4.5 mm. The specific energy consumption was very low (0.96-1.01 kWh/kg), considering the high values of the process temperature.

Unlike the industrial manufacturing process of LECA lightweight aggregate and the experimental processes presented above, that used conventional heating methods, the manufacturing technique described (Paunescu et al., 2019) was based on an heating unconventional method using microwave energy.

The unconventional method of microwave heating of materials has been known since the middle of the 20th century. Although it is a fast, economical and "clean" method, its application has remained in a very limited field, for drying and heating processes at low temperatures. Only in the last 10-15 years it has been experimentally found that several types of materials are suitable for microwave heating (organics, ceramics, metals, polymers, glasses, etc.) (Kharissova et al., 2010).

The current work aimed to produce a lightweight aggregate with high mechanical strength at the level of the best performing materials in the world using the microwave irradiation as an energy source.

2. METHODS AND MATERIALS

2.1. Methods

The method adopted for the production of lightweight aggregate is based on the technique of direct microwave heating of the material, pressed and protected with ceramic fiber mattresses introduced in the microwave oven (Figure 1b). The experimental scale tests were performed on a 0.8 kW-microwave oven (Figure 1a) of the type currently used in the household, adapted for operation at high temperature (up to 1200 °C). The microwave heating is completely different from the conventional heating because the thermal process is initiated in the core of material and then the heat is propagated to its peripheral areas. The material subjected to heating must be microwave susceptible, i.e. to be a dielectric material. Through the direct contact between the electromagnetic waves and the dielectric, it absorbs the waves in its entire volume and converts them into heat (Jones et al. 2002). Because the material itself is a heat generator, the heating process volumetrically and is very fast (Kitchen et al., 2014). An advantage of the microwave heating is the selective heating. Thus, only the material subjected to the thermal process is heated and not the massive components of the oven, as in the case of the conventional heating, resulting an important energy saving (Jones et al., 2002).





Fig. 1. Experimental microwave equipment a – microwave oven; b – positioning detail of the material protected with ceramic fiber in the oven

2.2. Materials

The materials used in experiment were coal ash and red clay from the recycled clay brick from the demolition of buildings, as raw material, and silicon carbide as a foaming agent.

The coal ash was purchased from the Paroşeni thermal power station. Its initial grain size was below 180 μm, being reduced after grinding and sieving below 63 μm. The chemical composition of coal ash was the following: 53.1% SiO₂; 23.7% Al₂O₃; 8.6%

Fe₂O₃; 7.9% CaO; 3.2% MgO and 3.5% Na₂O + K₂O.

The red clay waste was used after crushing and grinding it at a grain size below $80~\mu m$. Its chemical composition included: $60.6\%~SiO_2;~19.2\%~Al_2O_3;~8.1\%~Fe_2O_3;~2.6\%~CaO;~2.9\%~MgO;~1.2\%~Na_2O;~3.9\%~K_2O$ and $1.3\%~TiO_2$.

The silicon carbide was used at a grain size below 32 μ m, as purchased from the market.

Analyzing the chemical compositions of the two wastes used as raw material, it is found that they contain between 3.5 and 5.1% Na₂O + K₂O, which facilitates the microwave absorption (Rahaman, 2007; Kolberg and Roemer, 2001) and also Fe₂O₃between 8.1 and 8.6%, which allows efficient microwave heating of the material starting from the room temperature, although it contains significant proportions of SiO₂ and Al₂O₃ which are microwave transparent materials at low temperatures (Jones et al. 2002).

2.3. Characterization of the aggregate samples

The lightweight aggregate samples experimentally obtained by the sinteringfoaming process of the coal ash-clay waste were characterized by traditional analysis methods. The main physical, mechanical and morphological characteristics were: apparent porosity, thermal conductivity, density, compressive strength, water absorption and microstructural configuration of the samples. The apparent density was measured by the gravimetric method (Manual, 1999). The porosity was calculated by the comparison method (Anovitz and Cole, 2005) between the porous sample density and the density of the same material type in compact state obtained by melting followed by cooling to the room temperature. The determination method of the thermal conductivity (Bianchi-Janetti et al., 2015) consisted of measuring the thermal flow value that passes through a sample of standard dimensions (50 mm-thickness) placed between two metal plates. One of the plate was heated and protected with insulating material and the other was cooled. An own conception device was used to determine the compressive strength by developing an axial pressing force generated with a hydraulically operated piston. The last pressing force axially applied to the sample before to crack was considered the compressive strength value. The tested sample had a cylindrical shape with the diameter of 80 mm and the height of 70 mm. The water absorption of the porous sample was measured by the traditional method of its water immersion (ASTM D 570). The porous microstructure of the lightweight aggregate samples was identified with a Smartphone digital microscope.

Generally. for the physical characterization of aggregate the literature used the bulk density, characteristic of some samples with irregular shapes and relatively small dimensions. For this determination, obviously, it would have required a great number of pieces occupying a closed space with known volume, which should have been weighed. The ratio between mass and volume, which includes many free spaces between pieces, represents the bulk density. In the case of the experiments described in the paper, although several tests were carried out under similar conditions to ensure the veracity of the results, there were not enough samples that could be used to determine a bulk density. In this situation, the apparent density was measured and shown in the tables, although its value is clearly greater than the bulk density.

3. RESULTS AND DISCUSSION

3.1. Results

The experiments were conducted on a 0.8 kW-microwave oven in the experimental base of the Daily Sourcing & Research company of Bucharest. Four variants were constituted, including coal ash (between 47.0 and 52.2 wt.%) and clay brick waste (between 44.4 and 51.0 wt.%) as raw material, a foaming agent (silicon carbide) between 2.0 and 3.4 wt.% and water addition to facilitate the raw material pressing, keeping constant the proportion of 18 wt.%. The selection of raw material and

foaming agent was made based on previous experimental results from the literature and the own experience. The components distribution of the four variants is shown in Table 1.

Table 1. Composition of the experimental variants

Variant	Coal ash wt.%	Clay brick waste wt.%	Silicon carbide wt.%	Water addition wt.%
1	52.2	44.4	3.4	18.0
2	50.5	46.6	2.9	18.0
3	49.0	49.0	2.0	18.0
4	47.0	51.0	2.0	18.0

The functional parameters of the sintering-foaming process of the coal ash-clay waste and silicon carbide powder mixture are presented in Table 2.

Table 2. Functional parameters of the sintering-foaming process

Functional	y-roaming process Variant			
parameter	1	2	3	4
Dry/wet raw	448/	447/	447/	448/
material (g)	529	527	527	529
Process	1180	1170	1165	1160
temperature (°C)				
Heating time (min)	33	32	31.5	31
Average speed				
(°C/min)				
 heating 	35.2	35.9	36.3	36.8
- cooling	6.5	6.2	6.3	6.5
Expansion of				
material volume	190	180	160	150
(%)				
Lightweight				
aggregate amount	435	432	433	434
(g)				
Specific energy				
consumption	0.89	0.86	0.85	0.83
(kWh/kg)				

From the data presented in Table 2 it can be concluded that the direct microwave heating process is extremely energy efficient allowing to reach very high heating speeds (between 35.2 and 36.8 °C/min) and very short heating times (31-33 min), provided that the temperature of the sintering-foaming process is above 1160 °C. The higher weight proportion of coal ash influenced the foaming temperature value. The 20 °C difference between the process temperature in variant 1 (with 52.2 wt.% coal ash) and variant 4 (with

47 wt.% coal ash) was obvious. The very low level of the specific energy consumption value in all tested variants (between 0.83 and 0.89 kWh/kg) is remarkable, especially since the required process temperature was very high. The weight ratio of the foaming agent in the raw material mixture contributed to a higher expansion of the material volume in the case of variant 1 (with 3.4% silicon carbide) than variant 3 and 4 (with 2% silicon carbide). In the first case, the material expanded with 90% than its initial volume, compared to 50-60% in variants 3 and 4.

Table 3 shows physical and mechanical characteristics of the lightweight aggregate samples obtained by direct microwave heating.

Table 3. Physical and mechanical characteristics of lightweight aggregate samples

Characteristic	Variant				
	1	2	3	4	
Apparent density (g/cm³)	0.70	0.73	0.77	0.80	
Porosity (%)	68.2	66.8	65.0	63.6	
Compressive strength (MPa)	6.8	7.2	7.7	8.0	
Thermal conductivity (W/m·K)	0.129	0.135	0.140	0.148	
Water absorption (%)	7.0	6.6	5.7	5.2	
Pore size (mm)	0.9- 3.5	0.8- 3.0	0.8- 2.8	0.6- 2.5	

Analyzing the data from Table 3, it results that the aggregate samples experimentally manufactured by direct microwave heating correspond to the qualitative requirements of this type of material usable in the production of lightweight concrete.

The apparent density of the samples had relatively low values between 0.70 and 0.80 g/cm³. Compared with the values of other experimentally achieved aggregates measured as bulk density (0.77-0.86 g/cm³ [3, 7]), the samples obtained in these experiments are appropriate. Also, the porosity values between 63.6 and 68.2% were similar to the reference samples.

The compressive strength (between 6.8 and 8.0 MPa) was within the appropriate values of lightweight aggregates made by

conventional methods and the thermal conductivity had low values (0.129-0.148 $W/m\cdot K$) corresponding to the characteristics of good insulating materials. LECA aggregates industrially manufactured in the form of pellets had a thermal conductivity below 0.097 $W/m\cdot K$.

The experimentally-made aggregate was slightly water absorbent (between 5.2 and 7.0%), similar to other products of this type - although in [3] a value of only 4.2% was indicated, but the aggregate was in the form of pellets.

Pictures of the cross section of the lightweight aggregate samples are shown in Figure 2.

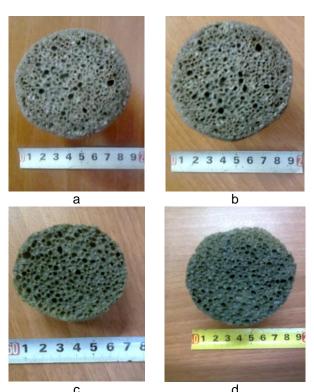


Fig. 2. Pictures of the lightweight aggregate samples
a – sample 1; b – sample 2; c – sample 3; d – sample 4

The microstructural characteristics of the four samples are presented in Table 3. The pore size decreases from 0.9-3.5 mm corresponding to sample 1 with the highest material expansion degree, up to 0.6-2.5 mm corresponding to the sample 4.

Images of the porous microstructure of the aggregate samples are presented in Figure 3.

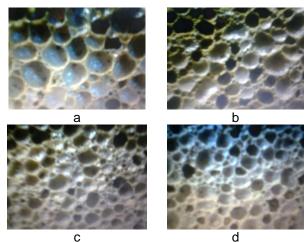


Fig. 3. Pictures of the porous microstructure of the lightweight aggregate samples a – sample 1; b – sample 2; c – sample 3; d – sample 4

According to the pictures in Figure 3, the pore distribution in the section of lightweight aggregate samples is homogeneous, indicating their insulating characteristics.

3.2. Discussion

The main advantage of using the direct microwave heating technique of the raw material powder mixture in the manufacturing process of the lightweight aggregate is the remarkable energy efficiency of this process, the conventional heating compared to techniques. According to the measurements carried out during the process, the heating speed reaches high values of over 35 °C/min. In addition, the microwave heating is an economical process because the heating is selective, without requiring additional energy consumption for heating the massive components of the oven. Also, the microwave heating process is an ecological process compared to the method of burning fossil fuels in combustion equipment.

It should be noted that the microwave is not considered a form of energy. It is only one way to convert an electromagnetic wave into heat by the direct contact with a microwave susceptible material or a powder mixture in which at least one of the components is microwave-susceptible.

4. CONCLUSION

A high-energy efficient heat treatment technique has been experimented to manufacture on a small scale a lightweight aggregate from an industrial by-product (coal ash) and a recycled waste from building demolition (clay brick waste).

The energy source was the microwave irradiation, directly applied on the raw material mixture (including also the foaming agent silicon carbide).

Four experimental variants containing fine mixture of coal ash (47-52.2 wt.%), clay brick waste (44.4-51 wt.%), silicon carbide (2-3.4 wt.%) and water (18 wt.%) were tested by heating in a 0.8 kW-microwave oven at temperatures between 1160 and 1180 °C.

The heating speed had very high values (35.2-36.8 °C/min) and the duration of the sintering-foaming process was very low (31-33 min.), considering the high level of the required temperature.

The products had relative low apparent densities (below $0.80~g/cm^3$), porosities with medium values (between 63.6~and~68.2%), low thermal conductivities (between $0.129~and~0.148~W/m\cdot K)$, water absorption in relative low limits (in the range 5.2-7%) and homogeneous porous structures.

The material was not pelletized, so that the product obtained was broken to adequate dimensions after cooling (10-80 mm).

The specific energy consumption of the sintering-foaming process was in very low limits (0.83-0.89 kWh/kg), theoretically well below the specific consumption of similar products manufactured by conventional methods, whose values are not provided in the literature.

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