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RECYCLED MASONRY RUBBLE USED AS RAW MATERIAL FOR MANUFACTURING LIGHTWEIGHT AGGREGATE BY AN UNCONVENTIONAL HEATING METHOD

BY

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Abstract. The paper presents an unconventional technique for manufacturing lightweight aggregate using microwave energy. The method is ecological, the basic raw material being masonry rubble, a waste from the building demolition. The originality of the work is the use of the method of direct microwave heating, which worldwide is industrially applied to a small extent and only in drying and heating at low temperature processes. The current work is a re-edition of the experiments performed by the same team of researchers in 2019, bringing a significant improvement of the degree of processing the raw material, which led to a reduction of its granulation. As an effect, the apparent density of the product was reduced to 0.40 - 0.54 g/cm^3, being also influenced the thermal conductivity reduced at 0.076 - 0.096 W/m·K and the porosity increased at 74.3 - 81 %. The compressive strength was not noticeably influenced, keeping in the range 5.5 - 7 MPa. The main advantage of the direct microwave heating method was the very low specific energy consumption (0.66 - 0.76 kWh/kg).

Keywords: energy consumption; lightweight aggregate; masonry rubble; raw material processing; unconventional technique.

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1. Introduction

Modern techniques for making light mortars and concretes involve the use of lightweight aggregates. Generally, they are made of natural resources (volcanic rocks). More recent techniques (Moeller *et al*., 2015) use natural waste (clay and slate) or industrial by-products (coal ash, sludge, etc.) for a synthetic manufacture at high temperatures, based on the release of gases into the raw material in a pyroplastic state, which favors the formation, spreading and blocking of gas bubbles in the viscous mass. Cooling this mass containing bubbles leads to the generation of a cellular structure with a sufficiently high voids volume to determine the light weight of the sintered and expanded material.

The masonry rubble is wastes from the demolition and rehabilitation of buildings. These wastes mainly contain concrete rubble, unmixed clay brick rubble and also calcium-silicate brick, aerated autoclaved concrete, precast concrete, other components as cement mortar, lime mortar, insulation, plasters, tiles and façade panels with high variation of their chemical composition (Moeller *et al*., 2015).

Waste from demolition and rehabilitation of buildings was a very important source of recycled raw material, especially after World War II. Germany and the United Kingdom were the world countries where this material resource reached its highest values. Also, currently the annual masonry rubble generation rate has high values of 70 million tons in the United Kingdom and 50 - 60 million tons in Germany (de Venny, 1999; Jakubcová *et al*., 2011).

According to the literature (Hammer *et al.*, 2000), the best-known technique for obtaining light aggregate is that of manufacturing lightweight expanded clay aggregates (LECA) by heat treatment of clay at 1200ºC in a rotary kiln. The manufacture of LECA began in the first decades of the last century in the United States. Several European countries (Denmark, Germany, the United Kingdom and Central and Eastern European countries) currently produce LECA. This aggregate type are manufactured in the form of pellets with dimensions below 25 mm, bulk density between 0.25-0.51 g/cm^3, thermal conductivity below 0.097 W/m·K, fire resistance and frost-thaw cycle resistance.

A method of manufacturing lightweight aggregates using only fly ash as solid waste is presented in the paper (Kayali, 2005). The sintering of fly ash based- raw material takes place at temperatures between 1090-1370ºC. At the exit of the oven, the sintered material is broken to the required dimensions. This aggregate type has a bulk density of 0.84 - 0.86 g/cm^3 and the water absorption is about 3 vol. %. The concrete made with lightweight fly ash aggregate is 21% lighter, has 15 - 21% higher mechanical strength and has a shrinkage value at least 30% lower than the traditional concrete.

Other experimental results in the manufacture of a sintered fly ash aggregate are presented in the paper (Gao *et al*., 2012). The mixture of the raw material was composed mainly of coal fly ash and, in lower proportions, clay, melted sludge, coal and paper-mill waste. The mixture was pelletized, pre-sintered at 700ºC for 25 min and sintered at 1250ºC for 20 min. The compressive strength of the aggregate was 7.8 MPa and the water absorption was 4.2 vol. %.

Manufacturing lightweight aggregate from masonry rubble is not a technique widely tested in the world. The paper (Moeller *et al.,* 2015) presents a manufacturing method using building materials waste containing concrete, natural aggregates, clay brick and other mineral components. Raw material processing included crushing, grinding (in a ball mill), pelletizing, firing and pellets stabilization (in a rotary kiln) operations. The foaming of the starting material was performed with silicon carbide as a foaming agent at 1160 - 1180ºC. The aggregate bulk density was 0.7 - 1 g/cm^3 and the material dimension was between 2 - 8 mm. The compressive strength of the lightweight aggregate was 4 MPa (by firing at 1160ºC) and 5 MPa (by firing at 1180ºC). The water absorption of material varied between 8 - 15.5 vol. % being significantly lower than that of lightweight expanded clay aggregate (LECA) (14 - 25.5 vol. %).

The literature (Jakubcová *et al.*, 2011) presents a technique for manufacturing lightweight aggregate from clay brick, mortar, plaster, lightweight concrete, usual concrete, sand lime brick and aerated concrete. The waste was crushed, ground in a ball mill and sieved at grain size below 100 μm. Silicon carbide was used as a foaming agent. The material was pelletized and heat treated in a rotary kiln supplied with fossil fuel. The optimal firing temperature was around 1185ºC leading to obtain the maximum material expansion under conditions of a silicon carbide dosage of 3%. By comparison, the quality of lightweight aggregate made of masonry rubble is almost similar to that of lightweight expanded clay aggregate (LECA).

All the methods of manufacturing lightweight aggregate presented above were performed by conventional heating techniques.

A team of researchers from the Romanian company Daily Sourcing & Research composed of authors of the current paper performed manufacturing tests of lightweight aggregate using an unconventional heating technique based on converting the microwave energy into heat (Paunescu *et al*., 2019). The peculiarities of a mixture of aluminosilicate materials (rubble masonry waste) including a high microwave susceptible foaming agent (silicon carbide) allowed the fast and economical direct heating of the mixture by microwave irradiation. The masonry rubble (85.6 - 90.8%) had an average composition including clay brick (48%), concrete (40%) and cement mortar (12%). Also, coal ash (between 4 - 9 wt. %) was an additional material and silicon carbide (between 3.5 - 5 wt. %) was used as a foaming agent. The required sintering temperature had values between 1168 - 1185ºC. Due to the direct microwave heating at very high rates (between 33.9 - 35.9ºC/min), the process time was relatively short (32 - 34 min). The lightweight aggregate had the apparent density between 0.75 - 0.98 g/cm^3, the thermal conductivity in the range 0.123 - 0.140 W/m·K, the compressive strength between 6 - 7.3 MPa, the water absorption between 12.9 - 14.6 vol. % 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), taking into account the high values of the process temperature.

Although known since the mid-20th century, the solid microwave heating has been industrially used only to a very small extent and only in drying and low temperature heating processes. The literature (Kharissova *et al.*, 2010) has recognized the energy efficiency of microwave heating compared to the conventional heating. In recent decades, it has been experimentally found that several types of materials (ceramics, organics, metals, glasses, polymers, etc.) are suitable for efficient microwave heating. However, the industrial application of this advanced technique is still delayed.

The objective of the current work was to improve the characteristics of the lightweight aggregate compared to those obtained in the previous experiment (Paunescu *et al.*, 2019) through a higher raw material processing. The heating technique used was also the direct microwave heating.

2. Methods and Materials

The process of manufacturing lightweight aggregate involves the expansion of the raw material heated to high temperature due to the release of a gas following a chemical reaction of decomposition or oxidation of the foaming agent. There must be a good correlation between the gas release temperature and the softening temperature of the raw material, so that the released gas meets a mass with adequate viscosity in which it can develop in the form of bubbles, but cannot leave it (Scarinci *et al.*, 2005).

By adopting silicon carbide (SiC) as a foaming agent, the chemical reaction that releases the foaming gas is:

$$SiC + 2O_2 = SiO_2 + CO_2 \quad (1)$$

The originality of the current work is the source of energy needed to heat the material in order to reach the temperature at which the foaming process takes place. The source adopted by the authors is represented by electromagnetic waves with the typical frequency of 2.45 GHz in contact with a microwave susceptibility material. This type of material (called dielectric) absorbs the electromagnetic energy in its entire volume and converts it into heat, the process being very fast (Jones *et al.*, 2002; Kitchen *et al.*, 2014).

Unlike the lightweight aggregate manufacturing methods mentioned above, which use pelletizing the raw material, in the experiment described in the current paper it was adopted the technique of loading a compact material mass in the oven and separating it into pieces by breaking at the end of the sintering/foaming process as in the case of industrial manufacture of foam glass gravel from glass waste (Geocell, 2017).

The microwave equipment for the manufacture of lightweight aggregate was a 0.8 kW-microwave oven of the type used in the household for food preparation, constructively adapted for operation at high temperature (up to 1200ºC). The mixture of finely ground materials was loaded wet and pressed into a mold and then released, being freely deposited on a bed of ceramic fiber mattresses at the base of the oven and covered with ceramic fiber mattresses. The purpose of this very intense thermal protection is to avoid the heat loss from the inside of the material subjected to heating to the outside. Unlike conventional heating, the microwave heating is initiated in the core of the material, where the energy of electromagnetic waves is converted into heat. The thermal level is maximum in the core of the material and propagates in the entire volume to the peripheral areas. That is why it is very important to conserve the thermal energy within the volume of the material protected with ceramic fiber mattresses. In this way, the unprotected metal walls of the oven do not heat up to more than 50 - 60ºC. The temperature control of the sintering/foaming process was performed by visualizing the upper surface of the irradiated material through 30 mm holes provided in the upper wall of the oven and in the upper layer of the ceramic fiber mattresses with a radiation pyrometer mounted above the oven at about 400 mm.

An overview of the experimental microwave equipment is shown in Fig. 1*a* (which was described in a previous article (Paunescu *et al*., 2021). A detail of the 0.8 kW-microwave oven during operation can be seen in Fig. 1*b* and the thermal protection of the material with ceramic fiber mattresses is shown in Fig. 1*c*.

Fig. 1 – Experimental microwave equipment,
a – overview of the microwave equipment; *b* – 0.8 kW-microwave oven (detail), *c* – ceramic fibber protection of the material.

The raw material used in the experiment was composed mainly of clay brick waste and usual concrete for the building in a weight proportion of about 90% (evenly distributed) and the rest (10%) cement mortar waste recycled from demolition waste. These proportions were approximately similar to those used in the previous experiment to microwave manufacture lightweight aggregate (Paunescu *et al*., 2019). The oxide composition of wastes is shown in Table 1.

Table 1
The Oxide Composition of Demolition Waste

Oxide composition (wt. %)	Clay brick	Usual concrete	Cement mortar
SiO_2	60.6	68.9	20.0
Al_2O_3	19.2	18.7	3.7
CaO	2.6	1.6	62.8
MgO	2.9	0.5	3.1
Fe_2O_3	8.1	6.5	3.1
Na_2O	1.2	0.2	0.1
K_2O	3.9	1.5	0.8
TiO_2	1.3	1.3	-
SO_3	-	-	3.2

The waste was crushed, ground together in a ball mill and sieved in the Romanian company Bilmetal Industries SRL Popesti-Leordeni (Ilfov) at a grain size below 100 μm, much lower than in the reference experiment (Paunescu *et al.*, 2019). In the raw material mixture coal fly ash purchased from the thermal power station Paroseni (Romania) was added, being processed in the ball mill at a grain size below 100 μm. The foaming agent was SiC at a fine granulation (below 40 μm) significant lower than those of this material used in the previous experiment (below 80 μm). The mixture containing demolition waste, coal fly ash and SiC was wetted with water (about 18 wt. %) to facilitate its cold pressing.

The experimental manufacture in microwave oven of lightweight aggregate from masonry rubble was performed adopting four variants where the weight ratio of coal fly ash varied between 4 - 7%, those of SiC in the range 3 - 4.5% and the masonry rubble proportion was between 88.5-93%. The adoption of the variation intervals of the components proportion of the starting mixture took into account the experimental results previously obtained. Table 2 presents the composition of materials used in the four experimental variants.

Table 2
The Composition of the Experimental Variants

Material composition	Variant 1 wt. %	Variant 2 wt. %	Variant 3 wt. %	Variant 4 wt. %
Masonry rubble	93.0	91.5	90.0	88.5
Coal fly ash	4.0	5.0	6.0	7.0
SiC	3.0	3.5	4.0	4.5
Water addition	18.0	18.0	18.0	18.0

3. Results and Discussion

The main functional parameters of the sintering/foaming process of manufacturing lightweight aggregate are shown in Table 3.

Table 3
Main Functional Parameters of the Manufacturing Process

Parameter	Variant 1	Variant 2	Variant 3	Variant 4
Raw material amount (dry/wet) (g)	480/566	480/566	480/566	480/566
Sintering/foaming temperature (ºC)	1160	1165	1172	1178
Heating time (min)	29.5	31	33	34
Average heating rate (ºC/min)	38.5	36.9	34.9	34.0
Average cooling rate (ºC/min)	6.5	6.4	6.5	6.4
Lightweight aggregate amount (g)	465	464	466	465
Specific energy consumption (kWh/kg)	0.66	0.70	0.74	0.76

According to the data in Table 3, the amount of raw material was kept constant in the four experimental variants: 480 g (dry), respectively 566 g (wet). The sintering/foaming temperature increased from 1160 ºC in the variant with 4% coal fly ash and 3% SiC, to 1178ºC in the variant with 7 % coal fly ash and 4.5% SiC. The higher proportion of coal fly ash influences the temperature of the foaming process increasing its value and the higher proportion of SiC favors the expansion process of the material. Due to the composition of raw material which predominantly includes SiO_2 and Al_2O_3 (being an aluminosilicate material), there is the possibility of the direct microwave heating with very high heating rate (over 34 C/min) without causing damage to the material structure during foaming, as happens in the case of silicate materials (*e.g.* glass). Consequently, the duration of the sintering/foaming process is short leading to obtaining very low specific energy consumptions (between 0.66-0.76 kWh/kg) below the values of conventional industrial processes.

The appearance of the lightweight aggregate samples in cross section is shown in Fig. 2. According to Fig. 2, the microstructure of the lightweight aggregate sample has a more inhomogeneous appearance with larger pores with the increase of the heating process duration from 29.5 to 34 min and the final process temperatures from 1160 to 1178ºC. The investigation of the physical, thermal, mechanical and morphological characteristics of the lightweight aggregate samples was performed using common methods of analysis.

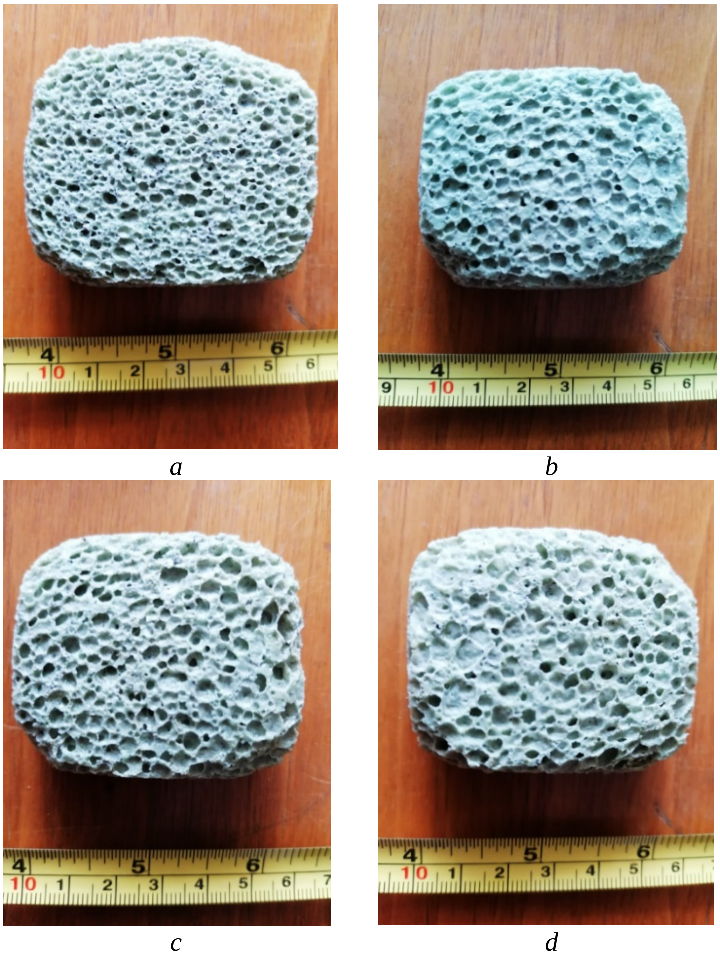


Fig. 2 – Appearance of cross section of lightweight aggregate samples,
a – variant 1; *b* – variant 2; *c* – variant 3; *d* – variant 4.

Thus, the gravimetric method (Manual, 1999) was used to determine the apparent density. The porosity was measured by the comparison method of porous material density and true density of compact material (Anovitz and Cole, 2005) and the compressive strength was measured with a TA.XTplus Texture Analyzer. The guarded-comparative-longitudinal heat flow method (ASTM E1225-04) was used to determine the thermal conductivity (Bianchi-Janetti *et al.*, 2015). The water absorption of the aggregate was measured by the water immersion method (ASTM D570) and the microstructural investigation of the porous products was carried out with an ASONA 100X Zoom Smartphone Digital Microscope.

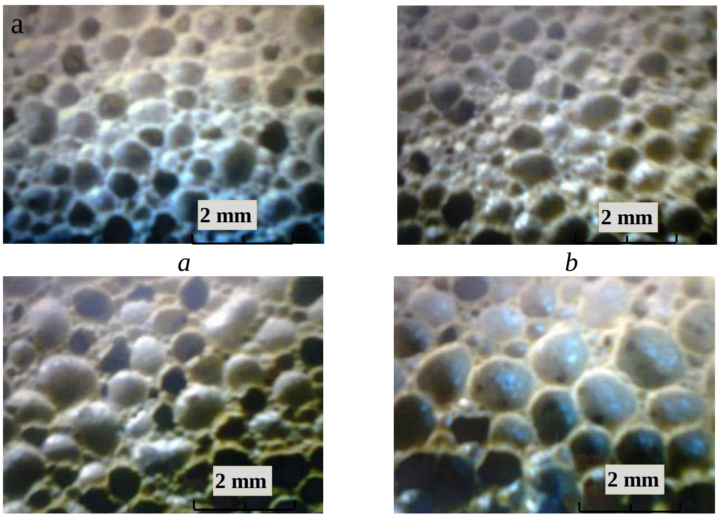
The results of measurements and determinations are centralized in Table 4.

Table 4
Physical, Thermal, Mechanical and Morphological Characteristics of the Aggregate Samples

Variant	Apparent density (g/cm^3)	Porosity (%)	Thermal conductivity (W/m·K)	Compressive strength (MPa)	Water absorption (vol. %)	Pore size (mm)
1	0.54	74.3	0.096	7.0	10.5	0.4-1.0
2	0.49	76.7	0.088	6.3	11.0	0.5-1.1
3	0.45	78.6	0.085	6.0	11.2	0.7-1.3
4	0.40	81.0	0.079	5.5	11.4	0.8-1.5

The data in Table 4 show the main effect of reducing the grain size of the raw material below 100 μm as well as the foaming agent below 40 μm by a higher mechanical processing. Thus, the apparent density of the expanded material was reduced from 0.77 - 0.98 g/cm^3 (Paunescu *et al.*, 2019) to 0.40 - 0.54 g/cm^3, the thermal conductivity decreased from 0.123 - 0.140 W/m·K (Paunescu *et al.*, 2019) to 0.079 - 0.096 W/m·K and porosity increased from 60.8 - 70.0% (Paunescu *et al.*, 2019) to 74.3 - 81.0%, indicating a significant improvement in the thermal insulation properties of the lightweight aggregate. The compressive strength changed insignificantly, having values between 5.5 - 7.0 MPa compared to 6.0 - 7.3 MPa in the reference experiment and the water absorption decreased to a small extent (10.5 - 11.4 vol. % compared to 12.9 - 14.3 vol. % in the reference experiment).

The microstructural configuration of the lightweight aggregate samples is presented in Fig. 3.



a *b* *c* *d*

Fig. 3 - Microstructural configuration of the aggregate samples, *a* – variant 1; *b* – variant 2; *c* – variant 3; *d* – variant 4.

The images in Fig. 3 indicate an acceptable microstructural homogeneity, the pore dimensions being included in the intervals shown in Table 4.

4. Conclusions

The objective of the current paper is to manufacture a lightweight aggregate by the unconventional method of microwave heat treatment, which allows to obtain lower energy consumption compared to conventional manufacturing techniques of the same product currently used in the world. The originality of the work is the use of this unconventional method whose industrial application worldwide is achieved only to a small extent and in thermal processes at low temperature.

Compared to a previous experiment in 2019 of the same team of researchers, in this case significant improvements have been made regarding the degree of processing of the raw material (masonry rubble and coal fly ash) and foaming agent (SiC), whose granulations have been much reduced.

The main effect of the change in the materials granulation was the decrease of the apparent density of lightweight aggregate at values between 0.40 - 0.54 g/cm^3 and, at the same time, of the thermal conductivity at 0.079 - 0.096 W/m·K and the increase of the product porosity value at 74.3 - 81.0%. The compressive strength of lightweight aggregate had no significant changes remaining in the range 5.5 - 7.0 MPa. The physical, thermal and mechanical characteristics of the aggregate are approximately similar to those industrially manufactured by conventional techniques.

The main advantage of the manufacturing method presented in the paper is the low level of specific energy consumption (between 0.66 - 0.76 kWh/kg) compared to the values of specific consumption of conventional industrial processes (over 0.80 kWh/kg).

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MOLOZ DE ZIDĂRIE RECICLAT UTILIZAT CA MATERIE PRIMĂ PENTRU FABRICAREA AGREGATULUI UȘOR PRINTR-O METODĂ DE ÎNCĂLZIRE NECONVENȚIONALĂ

(Rezumat)

Lucrarea prezintă o tehnică neconvențională de fabricare a agregatului ușor utilizând energia microundelor. Metoda este ecologică, materia primă de bază fiind moloz, un deșeu din demolarea clădirilor. Originalitatea lucrării este utilizarea metodei

încălzirii directe cu microunde, care pe plan mondial este aplicată industrial în mică măsură și numai în procese de uscare și de încălzire la temperatură redusă. Lucrarea curentă este o reeditare a experimentelor efectuate de aceeași echipă de cercetători în 2019, aducând suplimentar o îmbunătățire semnificativă a gradului de procesare a materiei prime, care a condus la reducerea granulației acesteia. Ca efect, densitatea aparentă a produsului a fost redusă la 0,40 - 0,54 g/cm^3, fiind influențate, de asemenea, conductivitatea termică redusă la 0,076 - 0,096 W/m·K și porozitatea crescută la 74,3 - 81%. Rezistența la compresiune nu a fost influențată in mod sesizabil, păstrându-se în intervalul 5,5 - 7 MPa. Principalul avantaj al metodei încălzirii directe cu microunde a fost consumul specific de energie foarte redus (0,66-0,76 kWh/kg).

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