

NONCONVENTIONAL NEW GENERATION OF ENVIRONMENTAL FRIENDLY AND ECONOMIC BUILDING MATERIALS: AERATED CONCRETE

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ABSTRACT: Experimentally, fine porosity-aerated concrete was made using the technique of corrosion the aluminum powder as a foaming agent in $Ca(OH)_2$ solution. The powder (below 10 µm) was obtained from recycled waste, melted by microwave irradiation and atomized by the contact with gaseous nitrogen jets in an original facility designed by authors. The making recipe of concrete is based on calcined gypsum as the main binding agent substituting the pollutant and non-economic cement, fly ash from industrial coal burning, perlite, silica fume, hydrated lime and distilled water. Aerated concrete with excellent thermal insulation properties (density of 601 kg/m^3, heat conductivity of 0.114 W/mK, and porosity of 75.2 %) was obtained. The compression strength was increased to 3.9 MPa by curing process with 0.3 bars-steam and 75 ºC for 4 hours and holding further 8 hours inside a sealed room heated to 80 ºC. The product belongs to the new generation of building materials made without CO_2 emission and with low energy consumption.
KEYWORDS: aerated concrete, aluminium powder, aqueous solution, calcined gypsum, steam curing.

1. INTRODUCTION

Since the first 20-30 years of the 20th century, the foundations for the manufacture of so-called aerated concrete were laid thanks to the Swede J. A. Erikson, known as the inventor of making principle by mixing limestone and ground slate [1]. A paper from the end of the last century [2] recorded that there are large reserves of slate waste in the world and there are conditions for its utilization as a raw material for the aerated concrete making. The modern manufacturing technique is based on mixtures including cement, lime, calcium sulfate (gypsum), fine sand, and fine particle of aluminum. Fine sand does not have the traditional role of aggregate. Aluminum powder, which plays a major role in foaming the mixture, reacts with sand or, in the case of using coal fly ash, with it generating a porous structure containing hydrogen bubbles. The role of adding the gypsum to cement mass is to slow down the hydration process of cement once it is mixed with water [3]. Thus, the cured concrete becomes a lightweight material. In the second half of the last century, it has been found that waste or industrial by-products can substitute lime or cement. In the current conditions, of the ecological and economic crisis [4], saving the cement is very beneficial, because it is an expensive and polluting raw material.

The advantages of applying aerated concrete in construction by comparison with traditional concrete are: keeping a comfortable indoor climate with lower costs, fire resistance, sound insulation, environmental sustainability [5].

More making procedures of aerated concrete are known depending on the binding component type (cement, limestone, combination of these materials, pulverized ash or granulated metallurgical slag), the rich-silica component (sand, coal fly ash, by-products of ferroalloys or ores making, etc.) and the fresh concrete curing processes [6, 7]. The main physical, heat and mechanical characteristics of aerated concrete are: density within the limits 200-1200 kg/m^3, heat conductivity between 0.06-0.40 W/mK and compression strength within the limits 0.4-15 MPa. The highest values of strength are obtained by curing with steam at high pressure of the fresh concrete.

Experimental manufacturing of non-autoclaved cellular concrete with mineral supplements was performed. Portland cement clinker, quartz sand (up to 90 % quartz), lime (above 80 % CaO and MgO) as raw materials and 5 wt.% diopside (56 % SiO_2 and 25 % CaO) or 7 wt.% wollastonite (53 % SiO_2 and 35 % CaO) as mineral additives were used. The role of mineral additives was the beneficial influence on the hydration of clinker and the generation of the structure of cured cement paste. The made product had density of 580 kg/m^3 (using diopside) and 600 kg/m^3 (using wollastonite), heat conductivity of 0.131 W/mK (using diopside) and 0.135 W/mK (using wollastonite) as well as compression strength of 3.3 MPa (with diopside) and 3.1 MPa (with wollastonite) [8].

In another work [9], fine sand as and metallurgical slag (50 % replacement of cement) as well as a polymeric sulfonate salt from organic components (within 0.2-2 % from the entire amount of cementitious material) led to increasing the compression resistance of concrete reaching 9.5 MPa (at the end of curing process of 7 days), but the foam concrete was too dense with the density around 1300 kg/m^3.

A non-autoclaved aerated concrete manufacturing technique was based on the use of phosphogypsum [10]. The optimal mixture composition content 15 % cement, 30 % granulated slag, 55 % phosphogypsum, 7 % quicklime, 1.6 % sodium sulfate, and 0.074 % aluminum powder. According to the authors, the ratio between water and solid components was 0.45. The temperature of the steam used in the curing process of fresh concrete was 90 ºC. The optimal foam concrete had the density of 600 kg/m^3 and the compression strength of 3.5 MPa. The experiment showed that phosphogypsum has the role to activate the making process. The proportion of aluminum powder was experimentally modified between 0.05-0.10 % and significantly influenced the reduction of material density due to growing the pore volume. The quicklime tested between 3-15 % strongly contributed to improving the compressive strength and sodium sulfate influenced the decrease of density and at the same time the increase of strength.

The major role of phosphogypsum (as an industrial by-product) on the compression resistance and setting time of concrete was highlighted in [11]. By the Taguchi method, it was shown that the cement, gypsum, and lime composition leads to significant increasing of mechanical properties of porous concrete. According to the authors of this paper, to obtain a product with density of 806 kg/m^3 and compression resistance of 2.09 MPa, the optimal ratios of the starting mixture should be 34 % Portland cement, 35 % gypsum and 10 % lime.

Different experimental results of manufacturing the cellular concrete are presented in [12]. The ground mixture representing the binding component included 30 % Portland cement, 30 % gypsum-based waste and 40 % ash-based waste. The amount of ash was 309 kg/m^3, while the amount of aluminum powder used as a foaming agent was 6.55 kg/m^3. The water quantity used in the making process was 371 kg/m^3 and the ratio between water and solid components was 0.74. The curing process of fresh concrete was carried out with steam at 34-36 ºC. The aerated concrete samples made under these conditions had the following characteristics: density between 580-950 kg/m^3, heat conductivity within the limits 0.17-0.24 W/mK, compression resistance between 3-8 MPa and the frost resistance coefficient after 50 experimental cycles was 0.86.

The reaction of aluminum particles in $Ca(OH)_2$ solution as an expanding agent of a ground mixture based on waste glass with the release of hydrogen in the form of bubbles at room temperature was previously tested by the authors of this work [13]. The procedure replaced the known methods of manufacturing cellular glass carried out at over 750 ºC) using foaming agents that release the required gas through a chemical reaction of decomposition or oxidation.

The same foaming technique based on aluminum particles in $Ca(OH)_2$ solution for the production of cellular glass was used by the research team mentioned above in the case of the experimental manufacture of aerated concrete [14]. The starting material mixture included calcined gypsum (70.7-78.8 %), $Ca(OH)_2$ (9.5-10.4 %), coal fly ash (3.4-5.1 %), perlite (10 %), silica fume (0.7-1.4 %), carboxymethyl cellulose-CMC (2 %), aluminum powder (3 %) and distilled water (25-35 %). The aerated concrete had density between 530-600 kg/m^3, porosity between 71.4-74.7 %, heat conductivity in the range of 0.129-0.184 W/mK, compression resistance within the limits 1.2-2.2 MPa and pore size below 4.5 mm.

Taking into account that a concrete structure with large pores has an unfavourable influence on the properties of cellular concrete, especially the compression resistance. Therefore, the objective of the current paper was focused on ensuring the appropriate relation between the values of density, heat conductivity and compression resistance as a result of processing a small pores-homogeneous structure uniformly distributed throughout the entire volume of the material. The original method of producing very fine aluminum particles (below 10 μm) through melting the recycled aluminum commercial packaging under the influence of microwave irradiation and then atomizing the molten through the interaction with nitrogen gas jets represents the main original character of the work.

2. MATERIALS AND METHODS

2.1 Materials

In principle, the manufacture of a monolithic material in ecological and economic conditions at the level of current international requirements involves the choice of appropriate materials that partially or totally exclude ordinary Portland cement. According to [15], calcined gypsum ($CaSO_4 \cdot 0.5H_2O$) is one of the binders with excellent physical-thermal features suitable for making the building concretes and, in addition, it is an environmental friendly material. Its association with an inorganic filler (perlite) is beneficial due to the ability of the filler to form homogeneous macrostructures with low pore size. An ultrafine powder (below 1 μm particle size) silica-rich (83-98 % SiO_2) with pozzolanic properties suitable for obtaining high-performance concrete is silica fume. Its addition to the concrete binder contributes to the increase of compression resistance, bond and abrasive resistance [16]. The lime addition as $Ca(OH)_2$ (obtained by the quicklime reaction with water) reacts with silica fume contributing to hardening the pozzolanic binder paste [17].

The use of fly ash (resulted as secondary product of energy industry) in the Portland cement concrete contributes to the increase of workability, mechanical strength, durability and improving the structural homogeneity of concrete [18]. To obtain moderate expansion of aerated concrete the fly ash proportion in this paper was limited below 11 wt. %.

The carboxymethyl cellulose (CMC) sodium salt added to the raw material composition had the role of foam stabilizer [19].

A major influence to obtaining the cellular concrete was played by the fine aluminum powder with the particle size below 10 μm as an agent supplying gas bubbles. The powder was made by atomizing with nitrogen jets the melt obtained by microwave heating a batch of recycled aluminum waste into a ceramic crucible made of silicon carbide (SiC). The process was carried out in the authors' experimental base and will be described below. The chemical composition of fly ash, perlite, and silica fume is indicated in Table 1.

Table 1. Chemical composition of solid materials

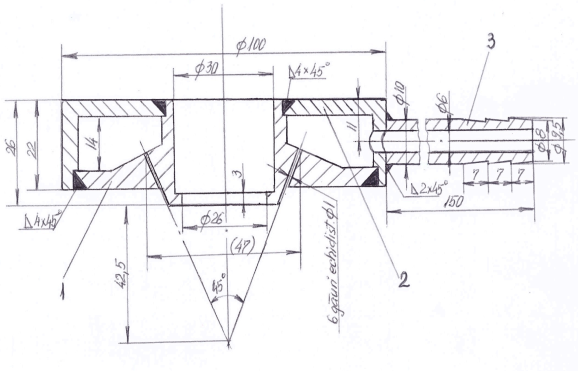
Composition	Fly ash (wt. %)	Perlite (wt. %)	Silica fume (wt. %)
SiO_2	46.3	71-74	85-98.7
Al_2O_3	23.9	1.2-1.5	< 2
Na_2O	6.0	3-4.1	3-5
K_2O	4.0	< 1.9	< 1.2
Fe_2O_3	8.4	0.4-1.9	< 1.9
MgO	1.4	0.2-0.8	< 2.0
CaO	8.0	0.5-1	< 2.4
Other oxides	-	-	< 4

2.2 Methods

The atomization installation of the aluminum waste melt in order to obtain the very fine powder of this metal consists of the atomization nozzle and the powder reception chamber (Figure 1). The atomization nozzle placed under the crucible containing the microwave-heated material allows free flow through a thin jet of molten metal, which intersects with several jets at 45 degrees of nitrogen gas at a high rate. The metal is pulverized and the fine particles fall into the metal reception chamber with double walls cooled intensively with water to avoid the formation of material agglomerations. The powder was captured at the base of installation at the end of operation. The method of producing the fine aluminum particles is the main originality element of the work.

a

b



c

Figure 1. Scheme and images of the atomization of molten aluminum
a, c – the atomization facility (image and constructive scheme);
b – image of aluminum particles.

In terms of chemistry, the method of obtaining the cellular concrete structure consists in releasing and blocking the gaseous hydrogen in the mixture mass in form of bubbles as a result of the chemical corrosion reactions on the aluminum powder surface in contact with the $Ca(OH)_2$ solution. Kaneshira et al. researched the application of this method [20] in order to produce hydrogen for primary energy. He has found that the ionized solution of $Ca(OH)_2$ containing Ca^{2+} and OH^- ions oxidizes the surface of aluminum particles to the form of Al_2O_3 turning it into aluminum hydroxide anion. This reacts with Ca^{2+} and water resulting so-named katoite [$Ca_3Al_2(OH)_{12}$] and hydrogen. The reaction occurs at ambient temperature and has the following form:

$$2Al + 3Ca(OH)_2 + 6H_2O = Ca_3Al_2(OH)_{12} + 3H_2 \quad (1)$$

Really, the process is more complex and takes place in several stages [20].

The process of preparing the material mixture has the following peculiarities. After dosing the amounts of the mixture components, their mixing and homogenization is carried out by mechanical stirring. Then, distilled water is poured over the dry homogenized mixture. During this time, the process of stirring the mixture is continued. The formation of the slurry allows its pouring into a metal mold with removable walls for freely expanding. Preparing the fresh cellular concrete takes place at 70-80 ºC obtained into a preheated radiant oven, where the vessel containing the slurry is introduced.

Modern industrial techniques for manufacturing autoclaved or non-autoclaved concrete use methods of hardening the fresh slurry using steam, hot air, keeping at room temperature, high pressure or atmospheric pressure, with process durations in general between 7-28 days in which only up to 1 day are subjected to the above treatments and the rest of the time is allocated for curing at room temperature. The curing process has the role of improving the concrete performance, especially mechanical strength [21]. The method adopted by the authors of this article had a particular character, the fresh material poured into metal molds being introduced into a hot room at 80 ºC kept constant. Steam was periodically used with the following parameters: 0.3 bars and 75 ºC. The complete maintaining time of concrete into the room was 12 hours and the steam blowing cumulated 4 hours.

2.3 Methods of characterizing the aerated concrete specimens

Generally, usual methods were applied in the process of determining the characteristics of aerated concrete samples. The use of standard PN-EN 12390-7:2011 allowed measuring the apparent density [22]. The porosity calculation was carried out based on estimating the fresh concrete density without pores and measuring the apparent density of porous specimen. The difference of the two values related to the value of concrete density without pores led to porosity determination (NE 012-99) [23]. The heat conductivity at room temperature was performed [24] according to SR EN ISO 8990:2002 and SR EN 1946-3: 2004. The compression resistance could be identified after hardening applying the axial pressing of cubic concrete specimens on an usual equipment used in the case of ordinary concrete (PN-EN 12390-3: 2011). The concrete water absorption (ASTM D570) was applied through its immersion under water for 24 hours. The microstructural aspect of concrete samples was examined with ASONA 100X Zoom Smartphone Digital Microscope.

3. RESULTS AND DISCUSSION

3.1. Results

As it appears from the list of materials chosen for the experimental manufacture of aerated concrete, traditional Portland cement (polluting and uneconomical) and ordinary aggregate are missing from this list. The new generation of concrete is based on calcined gypsum as the basic binder. Coal fly ash is already considered a very effective partial substitute for cement, hydrated lime contributes to the pozzolanic property of binder paste, silica fume is the pozzolanic additive in the binder, perlite as an additive in the binder reduces the cell dimension of concrete and CMC is a suitable foam stabilizer. Aluminum powder has a well-defined and major role in the foaming process.

Several testing variants were adopted, the chosen proportions of the materials being shown in Table 2.

Table 2. Composition of testing variants

Composition	**Variant (wt. %)**			
	1	2	.3	4
Calcined gypsum	82.4	81.3	78.3	74.6
Hydrated lime	5.1	5.5	7.4	7.9
Fly ash	3.5	4.9	6.9	11.0
Perlite	6.5	6.0	5.3	4.7
Silica fume	1.0	0.8	0.6	0.3
CMC	0.5	0.5	0.5	0.5
Aluminum	1.0	1,0	1.0	1.0
Water	41.3	40.7	39.4	37.5

Theoretically, fly ash, perlite, and silica fume influence the fineness and uniformity of the porous structure of aerated concrete. In the four variants, the proportion of fly ash increased from 3.5 to a maximum of 11.0 %. Consequently, perlite was reduced from 6.5 to 4.7 % and silica fume from 1.0 to 0.3 %. Also, the increase in the proportion of fly ash determined the moderate reduction of calcined gypsum from 82.4 to 74.6 %. Hydrated lime that forms the aqueous solution for corrosion reaction of aluminum particle was increased from 5.1 to 7.9 %, while CMC and aluminum powder were kept in all variants at constant values of 0.5 and 1.0 %, respectively.

Results of determining the features of aerated concrete specimens are shown in Table 3.

Table 3. Features of aerated concrete specimens

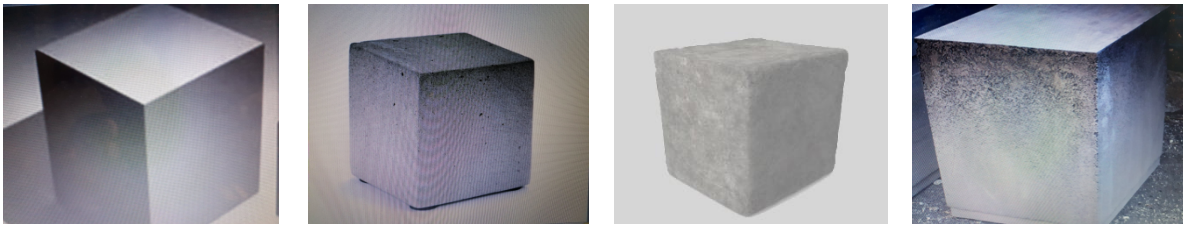
Feature	Variant			
	1	2	3	4
Apparent density (kg/m^3)	601	585	569	572
Porosity (%)	75.2	76.0	76.9	74.8
Heat conductivity (W/mK)	0.114	0.105	0.099	0.115
Compression strength (MPa)	3.9	3.7	3.6	3.3
Water absorption (vol. %)	3.6	3.2	2.8	2.8
Cell size (mm)	0.1-0.5	0.3-0.7	0.5-1.0	0.1-0.9

According to Table 3 containing the characteristics of the aerated concrete specimens produced in the four variants, it can be seen that apparent density, heat conductivity and porosity, i.e. those that indicate the heat-insulating properties specific to aerated concrete, have appropriate values for this objective. Thus, apparent density falls within the range of 569-601 kg/m^3, heat conductivity is between 0.099-0.115 W/mK and porosity has values within the limits of 74.8-76.9 %. By comparison with the values of the same characteristics obtained in the world and presented in the literature, they are generally similar. The water absorption in aerated concrete varied within normal limits (between 2.3-3.9 vol. %). The compression strength is a characteristic of aerated concrete strongly influenced by the curing conditions. In the case of the experiment presented in this paper, the strength reached acceptable values between 3.3-3.9 MPa, comparable to those of non-autoclaved concretes. Certainly, the aerated concrete steam cured in industrial autoclaves at high-pressure can reach much higher compression resistance values (11.6 MPa) by curing 6 hours in the autoclave and then 7 days in air [25].

During the experiment it was observed that by curing the fresh concrete the compressive strength can be significantly increased without the thermal conductivity changing visibly.

This effect is beneficial for the quality of aerated concrete, because usually, the increase of mechanical strength is equivalent to the increase of thermal conductivity corresponding to a denser structure of the concrete.

The appearance of aerated concrete specimens after the curing process is presented in Figure 2.

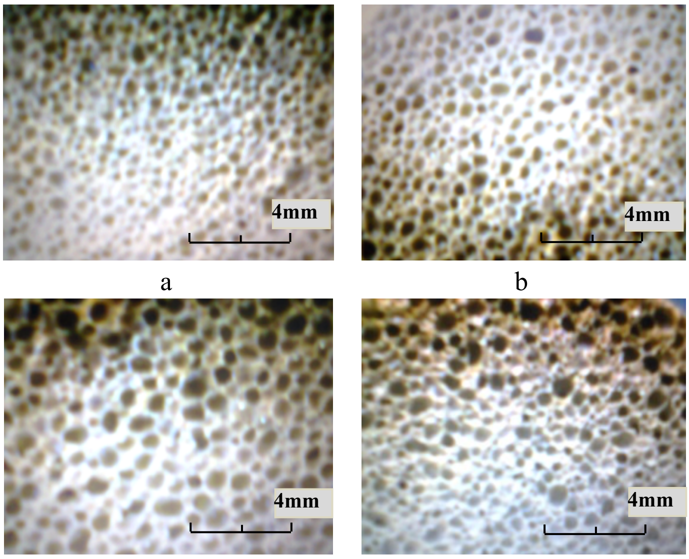


a b c d

Figure 2. Aspect of aerated concrete samples after the curing process
a – specimen 1; b – specimen 2; c – specimen 3; d – specimen 4.

Aerated concrete aspect shown in Figure 2 confirms that the increase of Al ratio as a foaming agent and also fly ash addition influenced the macrostructural homogeneity of material, dimension and uniformity of pore distribution, so that specimen 4 prepared with the highest fly ash ratio is characterized by the most disordered structure. Although its thermal insulation properties are excellent, its structure cannot guarantee the uniformity of thermal protection offered by this monolithic material. Instead, specimen 1 has the most homogeneous distribution of pores with very low sizes.

The microstructural homogeneous aspect of the monolithic material samples was verified by investigating the images obtained with the digital microscope in Figure 3.



a b c d

Figure 3: Microstructural aspects of aerated concrete samples
a – specimen 1; b – specimen 2; c – specimen 3; d – specimen 4.

Microstructures in Figure 3 show a progressive increase of pore size from variant 1 (a) to variant 3 (c). The sample representing variant 1 has the finnest microstructure (cell size within the limits 0.1-0.5 mm), while the specimen corresponding to variant 3 has a slightly larger microstructure (pore size between 0.5-1.0 mm). As mentioned above, the specimen corresponding to variant 4 has a relatively inhomogeneous microstructure (cell size within the limits 0.1-0.9 mm).

Taking into account the characteristics of cured aerated concrete specimens presented above, the making variant considered optimal was chosen variant 1. Prepared with 82.4 % calcined gypsum (the highest ratio), 5.1 % hydrated lime, 3.5 % fly ash (the lowest ratio), 6.5 % perlite, 1.0 % silica fume, 0.5 % CMC, 1.0 % aluminum powder, and 41.3 % distilled water, the aerated concrete had the following features: apparent density of 601 kg/m^3, porosity of 75.2 %, heat conductivity of 0.114 W/mK, compression resistance of 3.9 MPa and water aspirant of 3.6 vol. %.

3.2 Discussion

The manufacture of aerated concrete is a modern trend applicable in the field of building construction by significantly reducing the specific weight of concrete components (foundation, walls). On the other hand, the replacement of cement as a concrete binder has recently become a major requirement worldwide due to the fact that the industrial process of cement manufacturing is a great consumer of fossil fuel (fuel oil, natural gas, etc.) and also it is a generator of high greenhouse gas emissions (mainly CO_2). Under the conditions of the current ecological (climatic overheating of the planet) and energy crisis (reduction of the world's natural reserves of hydrocarbons) the replacement of cement with cheap natural materials whose processing is non-polluting and with various waste and industrial by-products also processed in non-polluting conditions is the optimal solution in the concrete making technique.

Like H_2O_2 and CaC_2, Al powder in $Ca(OH)_2$ liquid solution is the appropriate combination to produce the porous structure in concrete due to the hydrogen gas released in the mass of mixture and forming bubbles. This method of manufacturing aerated concrete was adopted by the authors of this work. The originality of the method was the fast and economic production of fine aluminum powder (below 10 µm) from recycled waste, melted by microwave irradiation in a silicon carbide crucible. The technique of nitrogen atomization of the aluminum melt was applied in an own conception installation, the fine metal particles being received at its base without the adhesion of several granules due to the intensively cooled walls with water.

The non-autoclaved curing of fresh concrete was achieved by an original adapted method using steam at 0.3 bars and 75 ºC for 4 hours and keeping the concrete specimens for another 8 hours inside an electrically heated room. The curing treatment of concrete allowed reaching relatively high values of compressive strength without affecting the excellent thermal insulation properties of aerated concrete.

4. CONCLUSION

The paper aimed at producing a new generation of environment friendly and economic building material: aerated concrete. The monolithic material was made under conditions in which the traditional Portland cement whose production involves great consumption of fossil fuel (fuel oil, natural gas, etc.) and generates high greenhouse gas emissions (mainly CO_2) was completely substituted with calcined gypsum (in high ratio around 80 %) having the main role of concrete binder. Industrial by-product of energy industry (coal fly ash) was also used as a partial replacer of cement additive, together with other additives that favour the formation of porous homogeneous concrete structure (perlite, silica fume, carboxymethyl cellulose). The chosen principle of making the aerated concrete is the elimination of a gaseous product by the chemical reaction of the peripheral surface of aluminium particles in contact with the liquid solution of $Ca(OH)_2$. The metal particles (below 10 μm) was made by the authors through the own original process of atomizing the recycled aluminum waste melted by fast and economic microwave irradiation. Atomizing the melt was carried out with gaseous nitrogen jets (to avoid the oxidation) in a sealed installation, which allowed the capture of metal particles without the adhesion of several granules between them. Curing the fresh concrete was performed through a own method of 0.3 bars and 75 ºC-steam blowing for 4 hours and then maintaining the specimens for 8 hours into a heated room at 80 ºC. The optimal variant of aerated concrete had density of 601 kg/m^3, heat conductivity of 0.114 W/mK, and compression resistance of 3.9 MPa.

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