



Lightweight gypsum composites: Design strategies for multi-functionality

Alena Vimmrová, Martin Keppert, Luboš Svoboda, Robert Černý*

Department of Materials Engineering and Chemistry, Faculty of Civil Engineering, Czech Technical University in Prague, Thákurova 7, 166 29 Prague 6, Czech Republic

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ABSTRACT

Two technological solutions for the preparation of lightweight gypsum composites are analyzed involving low-density foamed gypsum matrix with higher-density aggregates and higher-density matrix with low-density aggregates. In the design process, lightweight gypsum composites are considered as multi-functional materials which are supposed to have both load bearing function and thermal insulation capability. The design strategy with higher-density matrix and low-density aggregates appears to be more successful. The most favorable material resulting from this strategy contains expanded perlite in the amount of 5% of mass of gypsum. It has a bulk density of 547 kg m^{-3} , thermal conductivity of $0.12 \text{ W m}^{-1} \text{ K}^{-1}$ and compressive strength of 2.0 MPa. On the other hand, the designed composites with low-density foamed gypsum matrix and higher-density aggregates are found to have good thermal insulation properties but insufficient compressive strength. Nevertheless, this strategy is potentially viable if mechanical properties of the basic foamed-gypsum paste are improved, for instance by a moderate increase of bulk density.

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1. Introduction

Gypsum has been used as a construction material since ancient times. The oldest applications of gypsum plaster or gypsum binder were dated by archeologists to 7000 years B.C. [1]. The modern building industry can utilize not only natural gypsum, as commonly done in the past, but also forms of gypsum that appear as waste, secondary-raw material from various sources, such as phospho-gypsum [2,3], red gypsum [4,5], and flue-gas desulfurization gypsum [6,7].

The current application of gypsum in construction is relatively limited, despite the low price of raw material and general recognition of gypsum as environmental-friendly material. The most often use of gypsum in buildings is still in the form of interior plasters and plasterboards. However, the potential for gypsum applications is much wider. Gypsum blocks can possibly be used also in the exterior as load bearing elements if proper hydrophobization treatment is chosen [8]. Another prospective way is the development of lightweight gypsum which can be produced either as foamed-gypsum paste where the pore structure is formed by chemical additives [9,10] or using light aggregates [11,12].

In this paper, two basic technological solutions for production of lightweight gypsum composites are analyzed involving low-density foamed gypsum matrix with higher-density aggregates

and higher-density matrix with low-density aggregates. In the design process, lightweight gypsum composites are considered multi-functional materials having both load bearing function and thermal insulation capability.

2. Design strategies for multi-functional lightweight gypsum composites

In the first strategy low-density foamed-gypsum paste with low thermal conductivity (denoted as A-composition in what follows) was chosen as the basic material for the preparation of the composite. Crushed aluminum sulfate and calcium carbonate in the form of floated whiting which produce CO_2 gas when mixed together, $\text{Al}_2(\text{SO}_4)_3 + 3\text{CaCO}_3 + 3\text{H}_2\text{O} \rightarrow 2\text{Al}(\text{OH})_3 + 3\text{CaSO}_4 + 3\text{CO}_2$, were used as the foaming system. Further the mixture contained air entraining agent Silipon RN 7001 (producer Herkules GmbH, Germany), plasticizer Melment F10G (Stachema Kolín, Ltd.), polymer dispersion Vinnapas RI 551Z (Wacker Polymer Systems, Germany), cellulose Walocel MKX (Wolff Cellulosics, Germany) and retarding agent Retardan P (Tricosal GmbH, Germany). Polymer dispersion was used to enhance the mechanical properties, cellulose was supposed to stabilize the foam.

In the second strategy, the basic material was foamed-gypsum paste with higher-density (B-composition) where the achievement of higher strength was emphasized. Liquid acid agent prepared from aluminum sulfate and citric acid dissolved in water [13] was used as the acid foaming component. As the citric acid serves also as a retarder, adding of another retarding agent was not

* Corresponding author. Tel.: +420 224354429; fax: +420 224354446.

E-mail address: cernyr@fsv.cvut.cz (R. Černý).

Table 1
Composition of basic gypsum mixtures.

Mixture	Water/gypsum ratio	Citric acid	Aluminum sulfate	Calcium carbonate	Silipon RN 7001	Walocel MKX	Melment F10G	Vinnapas RI 551Z	Retardan P
% of gypsum powder mass									
G	0.93								0.1
A (lower-density paste)	1.07		11.36	2.87	0.02	0.34	0.53	6.25	0.1
B (higher-density paste)	0.53	3.67	7.33	2.34		0.2		8.35	

necessary. Using of air entraining agent and plasticizer turned out to be unnecessary as well.

A reference gypsum mixture denoted as G-composition was prepared to compare with the foamed mixtures. The detailed composition of basic gypsum mixtures used in the experiments is given in Table 1. The water/gypsum ratio was chosen in such a way that it complied with the requirement of normal consistence according to the Czech standard ČSN 72 2301 [14].

In the investigation of the above foamed-gypsum pastes, it was found that the generation of gas originating from the chemical reaction was not quite regular and sporadically large gas bubbles occurred in the material, their size being several orders of magnitude higher than the size of the prevailing pores (Fig. 1). This caused primarily considerable decrease of mechanical properties. However, the material was also not uniform enough which negatively affected its thermal insulation capabilities. Therefore, we decided to use fine granular fillers in gypsum mixtures, thus produce gypsum composites instead of gypsum paste.

The main motivation for producing fine-grained gypsum composites was our hypothesis that inert fine particles of the filler could act as nucleation sites during the chemical reaction of gypsum with water. In such a case, the reaction of gypsum particles with water would be initiated on the surface of the fine grains and the heterogeneous nucleation would take place in the system instead of the spontaneous, mainly homogeneous nucleation characteristic for gypsum pastes. The water available for the foaming agent would then be distributed more homogeneously over the whole volume of the material, thus the bubbles should be smaller.

The second, equally important, role of fillers in the composites was the adjustment of their thermal and mechanical properties through the directed variations of their density. Our intention was to get to the desired density from two opposite sides. The first was to begin with the A (lower-density) matrix and increase the density (and mechanical properties) of the composite by higher-density fillers. The second was to use the B (higher-density) matrix

and decrease the density (and thermal conductivity) of the composite by lower-density fillers. The amount of fillers appeared as crucial adjustment parameter in these procedures because it made possible to vary the density (and mechanical and thermal properties) of the composite in a wide range. The density of the matrix remained then only one of the decisive parameters affecting the properties of the composite.

The achievement of good mechanical and thermal properties of designed lightweight gypsum composites at the same time was the key factor for their multi-functionality. This opened a possibility for their use as materials of load bearing structures in a form of lightweight precast blocks with thermal insulation function. Also, they could be used as just thermal insulation boards if necessary. However, the newly designed materials retained some convenient properties characteristic for gypsum-based materials as well. Therefore, another possible application might be for renovation plasters where their high hygroscopicity can be utilized for the regulation of humidity in the interiors. The good fire resistance of gypsum offers then their use as fireproof plasters.

3. Materials and preparation technologies

For all compositions the flue-gas desulfurization gypsum from the power plant Mělník (producer Rigips) was used. As the fine higher-density fillers microsilica (Microsilica-Sioxid produced by OFZ Istebné, Slovakia), calcium carbonate (in high surplus compared to the above given values of “foaming carbonate”) and cement CEM I 42.5 were chosen. Expanded perlite EP 150 PB (grain size distribution in Table 2, loose bulk density 120 kg m^{-3} , produced by Perlit Praha, Czech Republic) was used as the light filler.

In the preparation of A-composition mixtures all solid components were mixed together at first and the dry mixture was added into the measured amount of water. Then, it was mixed by the laboratory mixer for 30 s at the low speed, than wiped off and mixed again for another 30 s at the same low speed. In the B-composition mixtures the dry components were mixed together with the gypsum powder. Measured amount of the acid agent was filled up to the needed volume of water and the dry mixture was added. Mixing was the same as in the case of A-composition mixtures.

The standard rectangular test prisms ($40 \times 40 \times 160 \text{ mm}$) were prepared from all mixtures, which were not compacted; only the surface was leveled by a knife. The specimens were dried at 50°C to the constant mass after demolding.

4. Experimental methods

Basic characterization of studied gypsum materials was done by means of their matrix density, bulk density and pore size distribution. Density of crushed samples was determined by helium

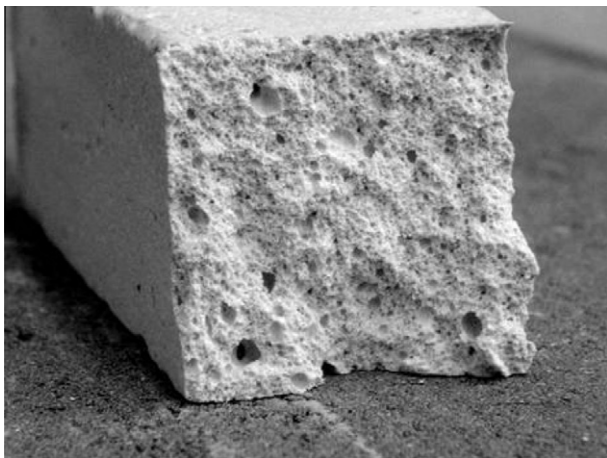


Fig. 1. Typical structure of the acid-foamed gypsum without any filler.

Table 2
Grain size distribution of expanded perlite.

Grain size (mm)	0–0.5	0.5–1.0	1.0–2.0	>2.0
Mass (%)	25–40	30–50	15–30	0–5

pycnometry (Pycnomatic ATC, Porotec, Germany). Small helium atoms penetrating well to the pores of studied samples ensure that the value of density measured by this method can be considered as the matrix density, ρ , of the porous material. The bulk density, ρ_b , was determined by weighing and size measurement of test prisms. Values of matrix density and bulk density were used for calculation of porosity and total specific pore volume.

Pore size distribution was determined by Mercury Intrusion Porosimetry (MIP, apparatus Pascal 140 + 440, Thermo Electron, Italy). Since the MIP, in the used measurement setup, detects only pores having radius smaller than 50 μm , volume of the larger pores was estimated as difference of the total pore volume and volume of pores detected by MIP. The possibility of error due to presence of large “ink bottle” pores inside the sample was reduced by size of samples for MIP (several pieces of size 2–3 mm, total sample mass was 0.3 g).

The measurement of bending strength was performed according to the Czech standard ČSN 72 2301 [14] on the $40 \times 40 \times 160$ mm prisms. The specimens were demolded 15 min after the final setting time and stored in the testing room. Every specimen was positioned in such a way that the sides that were horizontal during the preparation were in the vertical position during the test. The experiment was performed as a common three-point bending test. The distance of the supporting cylinders was 100 mm. The bending strength was calculated according to the standard evaluation procedure. The measurements were done at the time of 2 days after mixing.

Compressive strength was determined in accordance with the Czech standard ČSN 72 2301 [14] on the halves of the specimens left over after the bending tests. The lateral sides of the specimens which were in vertical position in the molds were in contact with the plates of the testing device. In this way, the imprecision of the geometry on the upper cut off side was not affecting negatively the experiment. The compressive strength was calculated as the ratio of the ultimate force and load area.

Thermal conductivity was measured using the commercial device ISOMET 2104 (Applied Precision, Ltd.) working on the impulse principle. The measurement was based on the analysis of the temperature response of the analyzed material to heat flow impulses. The heat flow was induced by electrical heating using a resistor heater having a direct thermal contact with the surface of the sample.

5. Experimental results and discussion

The characteristic material properties of foamed-gypsum pastes without any filler are presented in Table 3. While the matrix density of both A- and B-composition pastes was only about 5% lower than for the reference material G (which was mainly due to the specific additives such as polymer dispersion), the bulk density was greatly affected by the particular foaming procedures. For the A-composition it decreased more than three times and for B-composition by about 40%, as compared to the reference gypsum paste. Porosity of the plain gypsum (G) was found to be 59%; the dominating were pores with the radius of a few micrometers (Fig. 2). The adding of acid foaming agent to gypsum mixtures A

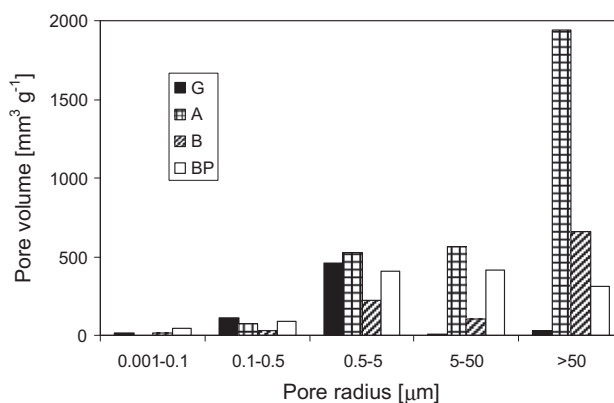


Fig. 2. Pore size distribution of gypsum-based materials.

and B caused significant change in the pore system structure. The total porosity increased to 87% and 69%, respectively. Moreover, the pore size distribution was changed and pores larger than 5 μm , bubbles with the radius above 50 μm in particular, prevailed in the pore system.

The foaming technologies caused the thermal conductivity (Table 3) to decrease five times in the case of A and two-and-half times in the case of B. However, the A-paste had the compressive strength 40 times lower than the reference G-paste, for B it was more than four times. Therefore, the basic foamed-gypsum pastes themselves could clearly not be considered as multi-functional materials. Adjustments of their mechanical and thermal properties using different types of fillers were necessary to match better the requirements placed for materials supposed to have both thermal insulation function and load bearing capability.

The addition of microsilica increased the bulk density of the foamed A-type composite by more than 50% at the microsilica amount of 15% of the gypsum mass (Fig. 3). The effect of cement was comparable to microsilica. CaCO_3 affected the bulk density increase in a less distinct way. The changes in compressive strength induced by the addition of higher-density fillers (Fig. 4) basically

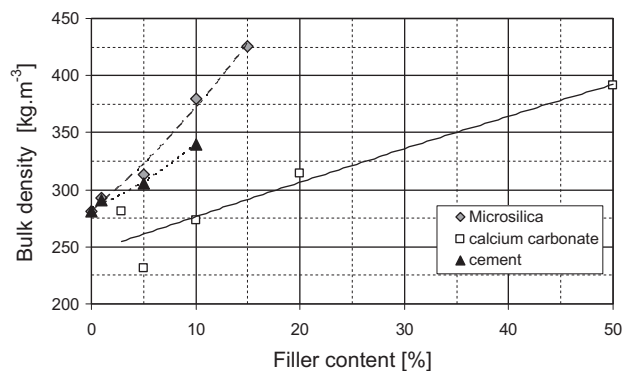


Fig. 3. Effect of higher-density fillers on the bulk density of foamed-gypsum composites (A-composition).

Table 3
Basic properties of the analyzed gypsum-based materials.

Material	Matrix density (kg m^{-3})	Bulk density (kg m^{-3})	Porosity (Vol.%)	Pore volume ($\text{m}^3 \text{kg}^{-1}$)	Thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)	Bending strength (MPa)	Compressive strength (MPa)
G	2314	945	59.2	0.62×10^{-3}	0.39	4.0	12.0
A	2207	281	87.3	3.10×10^{-3}	0.08	0.25	0.3
B	2193	661	69.4	1.04×10^{-3}	0.16	1.33	2.7
BP	1940	547	71.1	1.27×10^{-3}	0.12	1.44	2.0

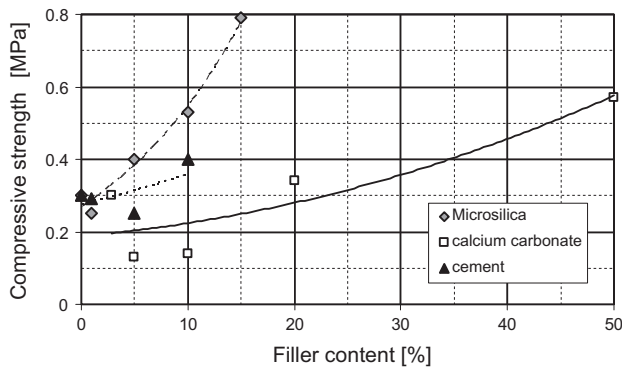


Fig. 4. Effect of higher-density fillers on the compressive strength of foamed-gypsum composites (A-composition).

followed the changes in bulk density; microsilica in the amount of 15% of gypsum mass increased the compressive strength almost three times, calcium carbonate at 50% of gypsum mass only two times.

The explanation of the more pronounced effect of microsilica on the compressive strength of the A-type composite as compared to the calcium carbonate (which is clearly an inert filler) may lie in the possible C–S–H structure development during the reaction between the calcium sulfate and microsilica as it was pointed out in [15]. On the other hand, the role of cement in the composite was not quite clear. The increase in compressive strength of the composite was distinct for 10% of gypsum mass only; thus it was difficult to tell if cement could or could not be considered as inert filler. In any case, the increase in compressive strength of A-composite was for all three used fillers not sufficient to consider the material suitable for load bearing purposes. It still remained mainly a thermal insulation material similar to the original A-composition gypsum paste.

The bulk density of B-type composite was affected by the addition of expanded perlite in a remarkable way. It decreased under 300 kg m^{-3} if perlite in the amount of 20% of gypsum mass was used (Fig. 5). However, the decrease of compressive strength to 0.3 MPa observed for 20% perlite addition was too high (Fig. 6). Apparently, the composite with addition of perlite in the amount of 5% of gypsum mass (it will be denoted as BP in what follows) could be considered as the best solution. Its bulk density was $\sim 20\%$ lower than for the basic B-paste but the compressive strength of 2 MPa was still in the acceptable range. The water/gypsum ratio of BP which led to the same consistence as for the basic mixtures given in Table 1 was 0.7.

The basic properties of the BP composite are shown in Table 3. The porosity was only slightly higher than for the basic B-mixture but the thermal conductivity decreased by 25% and the bending strength even slightly increased. The pore distribution of BP (Fig. 2) was much more homogeneous than of the B-paste. The amount of pores in the range over $50 \mu\text{m}$ decreased to about one half when compared with B but a significant increase of pore volume was observed in the ranges of $0.5\text{--}5 \mu\text{m}$ and $5\text{--}50 \mu\text{m}$. The differences in the pore structure of BP and B in the region of the largest pores could also be observed visually (Fig. 7). The structure of BP was characteristic by an almost complete absence of large cavity pores over 1 mm, whereas randomly distributed large cavities (1–5 mm) appeared in the structure of B. For a comparison the structure of reference gypsum G is given as well in Fig. 7, where practically no visible pores could be seen. The homogenization of the pore distribution of BP was a very positive outcome, in particular with respect to the thermal conductivity, water transport parameters and some mechanical properties of the composite.

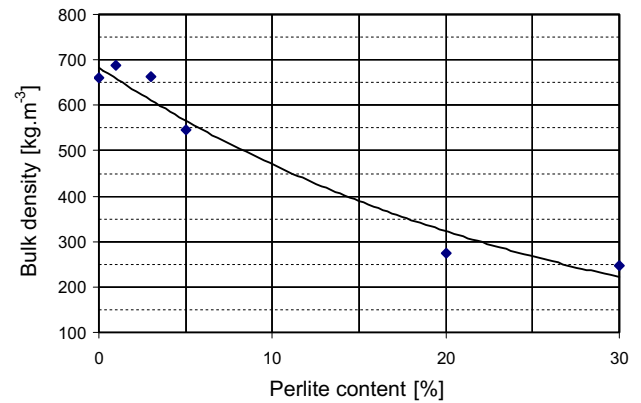


Fig. 5. Effect of perlite on the bulk density of foamed-gypsum composites (B-composition).

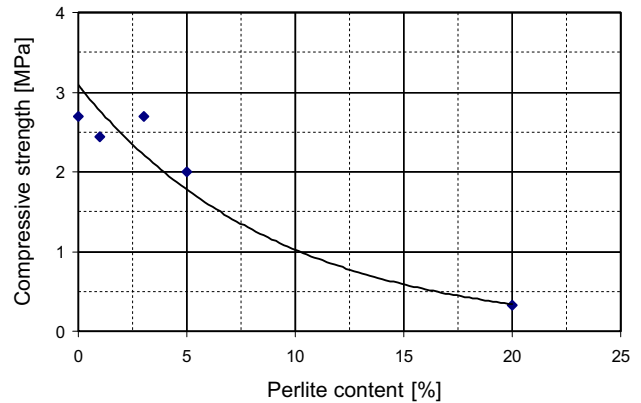


Fig. 6. Effect of perlite on the compressive strength of foamed-gypsum composites (B-composition).

The increase of bending strength and significant decrease of thermal conductivity (Table 3) can surely be attributed to the more regular pore structure of BP.

The lack of large pores and homogenization of pore distribution observed in the material BP confirmed our hypothesis from Section 2 that fine perlite particles probably acted as nucleation sites during the chemical reaction of gypsum with water. The water available for the foaming agent was then distributed more homogeneously over the whole volume of the material, and a higher number of smaller bubbles as compared with plain gypsum paste was generated.

A comparison with the properties of other foamed-gypsum materials can be done in a limited extent only because they were reported very rarely by other investigators in common sources. Colak [9] measured bulk densities and compressive strengths of several tens of foamed-gypsum mixes. For instance, one of his best foamed-gypsum materials with aluminum sulfate achieved the bulk density 694 kg m^{-3} and compressive strength 0.63 MPa, foamed gypsum with ammonium bicarbonate had the bulk density 756 kg m^{-3} and compressive strength 0.35 MPa. This is far worse than BP with the bulk density 547 kg m^{-3} and compressive strength 2.0 MPa.

Skujans et al. [10] reported several foamed-gypsum materials with the bulk densities from 200 to 700 kg m^{-3} . For instance, their material with the bulk density 500 kg m^{-3} achieved the thermal conductivity $0.16 \text{ W m}^{-1} \text{ K}^{-1}$ and compressive strength 0.7 MPa.

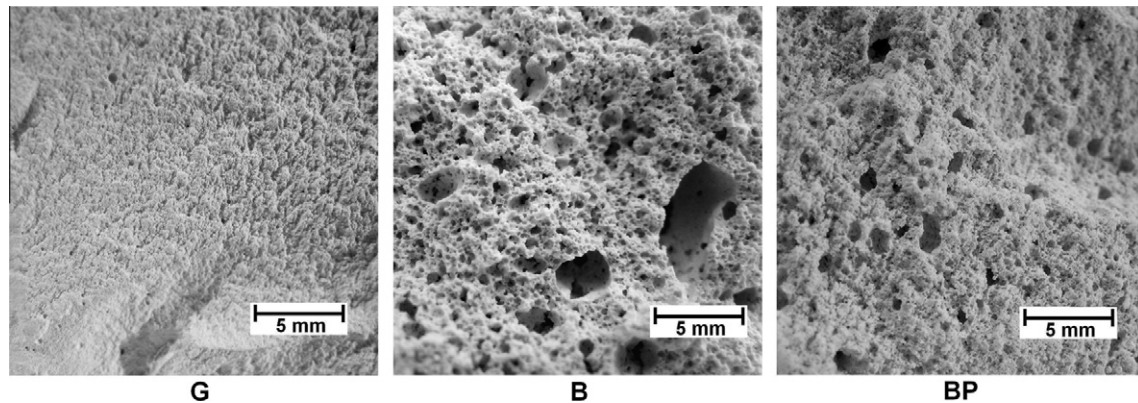


Fig. 7. Macrostructure of analyzed gypsum-based materials.

This was the same value of thermal conductivity as for the basic B-paste from this paper which had the bulk density 661 kg m^{-3} and compressive strength 2.7 MPa. The BP composite designed in this paper had the thermal conductivity $0.12 \text{ W m}^{-1} \text{ K}^{-1}$ and compressive strength 2.0 MPa with bulk density 547 kg m^{-3} so that both its mechanical and thermal insulation properties were significantly better as compared with the materials from [10].

The multi-functional BP composite from this paper can be considered as a possible competitor to the autoclaved aerated concrete (AAC). Therefore, it should be compared also with the available AAC products. We have chosen two characteristic AAC materials produced by Xella CZ for that purpose, namely P2-500 and P2-350 [16]. Ytong P2-500 ($125 \times 249 \times 599 \text{ mm}$) and Ytong lambda P2-350 PDK ($375 \times 249 \times 599 \text{ mm}$) are AAC blocks produced according to ČSN EN 771-4. Their supposed use is for non-bearing- (both P2-500 and P2-350) and load bearing (P2-350) walls, fireproof walls, and retention walls and linings in the interior.

The data given by the producer [16] shows that for instance the AAC material P2-500 has with slightly lower bulk density 500 kg m^{-3} the same thermal conductivity and compressive strength as BP but the material P2-350 achieves for 350 kg m^{-3} the thermal conductivity $0.085 \text{ W m}^{-1} \text{ K}^{-1}$ with the same compressive strength. Therefore, the commonly produced AAC materials have still better properties than the gypsum composites presented in this paper. However, it should be noted in this respect that AAC underwent extensive industrial research during the last several decades but lightweight gypsum composites were studied only rarely.

6. Conclusions

The experimental studies presented in the paper showed that lightweight gypsum composites have a potential to become multi-functional building materials if appropriate preparation technologies are chosen. Such lightweight materials can be used as thermal insulation boards, renovation- and fireproof plasters or in the form of load bearing lightweight precast blocks with thermal insulation function.

The main findings achieved in the investigations of lightweight gypsum composites can be summarized as follows:

- The fundamental problem of foamed-gypsum pastes was the formation of solitary large bubbles, which caused the irregular structure of the foam. The form of bubbles did not change by using surface active additives, which stabilize the foam, or by using polymer dispersion, which improves mechanical properties.
- The formation of large bubbles was successfully suppressed by adding fine granular fillers to the gypsum pastes.
- The design strategy for preparation of multi-functional gypsum-based composites with higher-density matrix and low-density aggregates (B-composition) turned out to be successful. The most favorable material BP resulting from this strategy contained expanded perlite in the amount of 5% of mass of gypsum. It achieved the thermal conductivity of $0.12 \text{ W m}^{-1} \text{ K}^{-1}$ and compressive strength of 2.0 MPa with bulk density equal to 547 kg m^{-3} so that its combination of mechanical and thermal insulation properties was significantly better as compared with similar materials designed by other investigators.
- The first advantage of the small perlite amount in the B-type composite was the more regular structure of the material without the large cavity pores over 1 mm. Presence of such large pores in material is undesirable, because they can deteriorate the physical properties such as the compressive strength, thermal conductivity and moisture transport parameters. The second advantage was the decrease of bulk density of the composite due to the low-density of expanded perlite which was achieved with still tolerable decrease of compressive strength.
- The design strategy with lower-density matrix and high-density aggregates (A-composition) was found only partially successful. All three tested fillers, namely microsilica, cement and calcium carbonate, reduced significantly the development of large bubbles in the pore system as compared to plain foamed gypsum. However, the increase in compressive strength of the A-composite was (for all three fillers considered) not sufficient to consider the material suitable for load bearing purposes. It still remained mainly a thermal insulation material similar to the original A-composition gypsum paste.
- Among the three fillers used in the preparation of A-composites, microsilica was the most promising solution, probably due to the C–S–H structure development during the reaction with calcium sulfate. On the other hand, for calcium carbonate which was apparently an inert filler the increase of strength upon the filler concentration was far less steep. The strength improvement achieved with cement was somewhere in between; it was not quite clear if cement was or was not an inert filler.
- Despite the only partial success with the preparation of gypsum-based composites using the A-composition approach, this strategy is potentially viable if the mechanical properties of the basic foamed-gypsum paste are improved, for instance by a moderate increase of bulk density.

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