Influence of fillers on structure and behaviour of gypsum mortars

M Doleželová¹, J Krejsová¹, L Scheinherrová¹, A Vimmrová¹*

¹Czech Technical University in Prague, Faculty of Civil Engineering, 166 29 Prague 6, Czech Republic

*E-mail: vimmrova@fsv.cvut.cz

Abstract. Gypsum mortars with different types of lightweight fillers were studied. The importance of gypsum as one of the most environmentally friendly building binder increases nowadays and new ways of its utilization are searched. Using of fillers in the gypsum is generally not necessary, because gypsum does not shrink, but they can be used for economic reasons or to improve the thermal properties or fire resistance of the gypsum material. We studied the structure and properties of the gypsum mortars with three types of lightweight fillers (expanded perlite, expanded clay aggregate, recycled PUR) and compared them with the gypsum mortar with siliceous sand. The SEM analyses were performed together with the mechanical tests. The roughness of the fillers surface and its influence on the structure and properties was studied by confocal laser scanning microscopy. It was found, that the type of the filler has principal impact on the microstructure of gypsum mortars and their properties.

1. Introduction

The significance of gypsum, as one of the most environmentally friendly building binders is currently increasing and new ways of its utilization are searched. Generally, the fillers are not necessary in the gypsum-based materials, because gypsum (contrary to cement-based materials) does not shrink during setting [1]. Nevertheless, the fillers can be used to improve e.g. the fire-resistant properties of gypsum, because the gypsum paste without fillers shrinks significantly at high temperatures [2]. Lightweight fillers can be used in order to improve the thermal resistance of the gypsum materials.

As a lightweight filler perlite [3] or vermiculite [4] are used most often, also expanded polystyrene, either new [5] or recycled [6] can be used. Other waste polymer materials were tested, e.g. polyurethane foam from automobile and construction industry [7] or ground rubber from used tires [8] or from pipe foam insulation [9]. Mostly the impact of the filler on the properties of gypsum material was studied and all authors state, that by addition of any lightweight aggregate the thermal properties of gypsum material were improved, but the mechanical properties usually decreased significantly. The impact of the filler on the structure of the materials was not studied often and also direct comparison of different types of fillers used in the same gypsum material was not performed yet.

We studied three types of fine lightweight aggregates – perlite, ceramic aggregate and waste ground polyurethane and their influence on the structure and properties of gypsum mortar. The results were compared with the gypsum mortar containing standard sand.
2. Materials

All mortars were prepared from the commercial gypsum plaster (calcium sulfate hemihydrate, CaSO\(_4\)-1/2H\(_2\)O), calcined from flue gas desulfurization product (CaSO\(_4\)-2H\(_2\)O), type A2 according ČSN EN 13 279-1:2009 [10] (producer Saint-Gobain Construction Products CZ, branch RIGIPS, Czech Republic).

As a lightweight aggregate the expanded perlite EP 150 (producer PERLIT, Czech Rep.), expanded clay aggregate Liapor (producer LIAS Vintířov, Czech Rep.) and crushed waste polyurethane foam from used white goods (producer DAXNER Technology, Czech Rep.) were used. The standardized sand CEN, ČSN EN 196-1 [11] was employed for the preparation of the reference material. The mineral composition of raw materials (when possible), obtained by XRD analysis is given in the Table 1. The granulometry of the aggregates can be seen in Figure 1 and their bulk densities (provided by the producers) are in Table 2. Set retarder Retardan-200 P (producer SIKA, Germany) was used in all mortars. There can be seen, that even if the maximal size of particles in all aggregates was declared as 2 mm, the crushed PUR has the finest granulometry with particles smaller than 0.5 mm, while expanded perlite has most of the particles between 0.5 and 2 mm.

### Table 1. Mineral composition of raw materials.

| Mineral          | Gypsum [wt. %] | Standard sand | Expanded clay | Expanded perlite* |
|------------------|----------------|---------------|---------------|------------------|
| Bassanite        | CaSO\(_4\)-1/2H\(_2\)O | 89            |               |                  |
| Anhydrite        | CaSO\(_4\)   | 6             |               |                  |
| Calcite          | CaCO\(_3\)   | 4             |               |                  |
| Muscovite        | Ka\(_2\)(AlSi\(_3\)O\(_{10}\))(OH)\(_2\) | 1            |               | traces           |
| Quartz           | SiO\(_2\)    | 100           | 6             |                  |
| Mullite          | Al\(_6\)Si\(_3\)O\(_{13}\) | 38           |               |                  |
| Anorthite        | CaAl\(_2\)Si\(_2\)O\(_8\) | 23           |               |                  |
| Hercynite        | Fe\(^{2+}\)Al\(_2\)O\(_4\) | 21           |               |                  |
| Diopside         | CaMgSi\(_2\)O\(_6\) | 6            |               |                  |
| Hematite         | FeO\(_2\)    | 4             |               |                  |
| Magnesite        | MgCO\(_3\)   | 2             |               |                  |

* Glassy material – mostly amorphous

### Table 2. Bulk density of used aggregates.

| Aggregate     | Standard sand [kg/m\(^3\)] | Expanded clay | Expanded perlite | Crushed PUR foam |
|---------------|-----------------------------|---------------|------------------|------------------|
| Bulk density  | 2575                        | 1050          | 80               | 180              |
2.1. Composition and preparation of mortars.

The composition of the mortars was designed so that the volume of gypsum and aggregate was the same in all mortars. As an initial composition the reference mortar GR with standard sand was chosen and its composition was designed according ČSN EN 196-1 [11]. The mass ratio between the gypsum powder and standard sand was 1: 3. Amounts of 450 g of gypsum and 1350 g of sand were used for one batch of reference mortar. The mass of each aggregate in other mortars was calculated from its bulk density $\rho_V$ and the volume of standard sand $V_R$ in mortar GR (Eq. 1) in order to maintain the same volume of aggregate in all mortars.

$$m = \rho_V \cdot V_R$$

The amount of water was determined for a flow tests diameter value of $165 \pm 5$ mm and there can be seen, that the mortar with PUR needed substantially higher amount of water than mortars with inorganic aggregates. The amount of retarding agent was 0.02% from dry gypsum weight in all mortars. Composition of tested materials is in Table 3.

**Table 3.** Composition of tested mortars.

| Material | Aggregate type       | Gypsum [g/batch] | Aggregate [g/batch] | Retardant [ml/batch] | Water [ml/batch] |
|----------|----------------------|------------------|---------------------|----------------------|------------------|
| GR       | standard sand        | 450              | 1350.0              | 0.09                 | 270              |
| GEC      | expanded clay        | 450              | 550.5               | 0.09                 | 320              |
| GEP      | expanded perlite     | 450              | 41.9                | 0.09                 | 260              |
| GPUR     | crushed PUR foam     | 450              | 94.4                | 0.09                 | 400              |

![Figure 1. Particle size distribution of used aggregates.](image-url)
The materials were prepared according to ČSN EN 13454-2 [12]. First, the required amount of water was poured into a bowl. Then dry gypsum was mixed with the retarding agent and dry mixture was poured into the water. The bowl was immediately placed in an automatic mixer. The total mixing time was 4 minutes, which included mixing for 30 s at low speed, 30 s of adding aggregates at low speed, 30 s mixing at high speed, pause 90 s during which the mixture was manually wiped from the bowl wall and finally mixing for 60 s at high speed. Then the gypsum mixture was poured into a mold and was firstly manually and then mechanically compacted. From each material the three prismatic samples 40 x 40 x 160 mm were prepared. The surfaces of the samples were levelled using a metal knife. The hardened samples were tipped out of the molds 60 - 90 min later and stored at 20 ± 5 °C and relative humidity of 50 ± 5%.

3. Methods
The particle size distribution was determined according to EN 933-2 [13]. Standard sieves with apertures of 0.063 mm, 0.09 mm, 0.125 mm, 0.25 mm, 0.5 mm, 1.0 mm, 2.0 mm and 4.0 mm were used.

The microstructural morphology of the gypsum mortars and surfaces of the aggregate grains was studied by scanning electron microscopy (SEM) using a Phenom XL electron microscope. The samples were dried and they were not coated or polished.

The grain and fracture surface roughness was determined by confocal laser scanning microscopy (CLSM) as the three-dimensional arithmetical mean roughness value SRa [µm] by the Phenom XL. The three-dimensional roughness is an analogy to the arithmetical mean deviation of the profile Ra (two-dimensional roughness) according to ISO 468 [14]. The SRa value of the typical grain surface of each aggregate was determined according Eq. 2.

$$SRa = \frac{1}{l_1 l_2} \int_{0}^{l_1} \int_{0}^{l_2} f(x, y) dx dy$$

where f(x,y) is height of the assessed profile and l1, l2 are measured lengths.

The bulk density of mortars was calculated from the mass and dimensions as a ratio of mass and volume. The dimensions of samples were measured by the digital calliper and their mass was determined by weighing. Volume of samples was calculated from dimensions.

The flexural and compressive strength were determined according EN 13454-2 [12] on standard test samples. The experiment was performed as a common three-point bending test. The measurements were carried out 7 days after mixing. The compressive strength was measured on the halves of the specimens left over from the bending test.

4. Results and Discussions
The shape and quality of the surface of the aggregate grains were studied by the SEM. From each aggregate several grains was chosen and the surface roughness SRa was measured 10 times for each aggregate at 20 µm wavelength and the mean value was calculated. The surface of typical grain of each aggregate type and the mean surface roughness are given in the Table 4. There can be seen that the roughest surface has the standard sand, because it was crushed at production process in order to obtain the required particle size. The crushed PUR differs significantly from other materials and it has also the smoothest surface of all aggregates. The values of the surface roughness correspond to the visual observations. There can be also seen, that particles of expanded clay and perlite are very porous, while sand and PUR particles have not any pores inside.
Table 4. Shape and surface roughness of aggregates.

| Aggregate           | standard sand | expanded clay | expanded perlite | crushed PUR |
|---------------------|---------------|---------------|------------------|-------------|
| SEM 500 x magnif.   | ![SEM image](image1.png) | ![SEM image](image2.png) | ![SEM image](image3.png) | ![SEM image](image4.png) |
| SRa [μm]           | 0.591         | 0.557         | 0.474            | 0.351       |

SEM images of the mortars with the different fillers are in the Figure 2. The images taken at 250x magnification show the overall structure of the mortars. There can be seen that the microstructures of mortars differs significantly. While in the mortars with well-defined aggregates (i.e. sand and expanded clay) the boundary between the aggregate particles and gypsum can be seen distinctly, in the mortars with less regular particles (perlite and PUR) the crystals of gypsum grows into the surface of the particles. At magnification 3000 x can be also seen, that the size of gypsum crystals in particular mortars differs. The largest gypsum crystals are in the mortar with PUR (length approx. 13 μm, thickness 2 μm) and they are separated from the particle surface and loosely packed and distinct interfacial transition zone (ITZ) is formed. The smallest particles can be seen in the mortar with expanded clay (length approx. 8 μm, thickness 1 μm) and the structure is less porous. The densest structure is formed in the mortar with perlite, where the gypsum crystals are shorter (length approx. 10 μm, thickness 1.5 μm) and they are not as smooth as crystals in other mortars. There can be said, that the biggest crystals and the most distinct ITZ were formed in the mortars with nonporous particles, regardless of the surface roughness. Also, the amount of water in the mixture did not play any important role in the crystal formation.

The values of bulk density, strength and roughness of fractured surface are given in Table 5. The fracture surface roughness was measured on the broken surface after bending test at the wavelength 50 μm.

Table 5. Physical properties of mortars.

|                        | standard sand | expanded clay | expanded perlite | crushed PUR |
|------------------------|---------------|---------------|------------------|-------------|
| compressive strength   | [MPa]         | 12.7          | 12.5             | 9.4         | 2.9         |
| flexural strength      | [MPa]         | 5.9           | 4.3              | 3.9         | 1.8         |
| bulk density           | [kg/m³]       | 1886          | 1107             | 647         | 838         |
| fracture surface roughness | [μm]      | 1.46          | 1.72             | 1.74        | 1.47        |
| Aggregate       | 250x                                                                 | 3000x                                                                 | Structure                                                                 |
|-----------------|----------------------------------------------------------------------|----------------------------------------------------------------------|----------------------------------------------------------------------------|
| standard sand   | ![Image](image1.png)                                                  | ![Image](image2.png)                                                  | · large, well formed crystals  
                   |                                                                     |                                                                     | · smooth crystals  
                   |                                                                     |                                                                     | · porous structure  
                   |                                                                     |                                                                     | · distinct ITZ       |
| expanded clay   | ![Image](image3.png)                                                  | ![Image](image4.png)                                                  | · smallest, well formed crystals  
                   |                                                                     |                                                                     | · less porous structure  
                   |                                                                     |                                                                     | · less distinct ITZ   |
| expanded perlite| ![Image](image5.png)                                                  | ![Image](image6.png)                                                  | · medium size, frayed crystals  
                   |                                                                     |                                                                     | · densely packed       |
                   |                                                                     |                                                                     | · no visible ITZ                                                     |
| crushed PUR     | ![Image](image7.png)                                                  | ![Image](image8.png)                                                  | · largest crystals             
                   |                                                                     |                                                                     | · distinct ITZ         |

**Figure 2.** The SEM images of microstructure of mortars.
There can be seen, that the values of strength are related to the surface roughness of aggregate particles (Fig.3). The rougher the particles are the higher the strength is. This phenomenon is known in the materials with cement binder [15] and was observed by the authors also in the gypsum mortars [16]. It can be seen that even if the bulk density of mortar with expanded clay decreased to 60% of the bulk density of mortar with standard sand, the compressive strength stayed nearly the same and fracture strength decreased 14% only. On the opposite side the strength of mortar with PUR decreased significantly, because of very smooth PUR particles.

Unlike the strength the fracture surface roughness does not correspond with the roughness of particles. The fracture surface roughness depends more on the porosity of particles. It can be explain by the fact that the nonporous particles are stronger and therefore not easily broken and the fracture line goes through the gypsum paste only, which is relatively smooth. In the mortars with weaker porous particles the fracture line goes through the broken particles also and these broken particles has rougher surface than gypsum paste and therefore the fracture surface roughness is bigger.

![Figure 3](image.png)

**Figure 3.** Relationship between the particle surface roughness and strength of mortars.

5. Conclusions
The influence of the different types of the lightweight aggregates was studied. It was found, that the structure of gypsum was changed by the presence of different types of particles and the type of particles had also the influence on the physical properties.

There conclusions can be drawn:
- The rougher the surface of aggregate particles is, the higher the strength of gypsum mortars.
- The size and shape of gypsum crystals in the vicinity of particles depends on the surface roughness of aggregates. In the mortars with smoother particles the gypsum crystals are larger and better defined and there is more pores between them.
- The fracture surface roughness depends on the strength of the aggregate particles. Stronger particles do not break during the bending and the fracture goes through the smoother gypsum only, while the weak porous particles breaks and the fracture goes through the rougher particles also.

Acknowledgments
This research was supported by the Czech Science Foundation, Project No. 19-08605S.
References

[1] Karni J, Karni E. 1995 Gypsum in construction: origin and properties. Mater Struct. 28 92 doi:10.1007/bf02473176

[2] Doleželová M, Scheinherrová L, Krejsová J, Vimmrová A. 2018 Effect of high temperatures on gypsum-based composites. Constr Build Mater. 168 82 doi:10.1016/j.conbuildmat.2018.02.101

[3] Demir I, Serhat Baspinar M. 2008 Effect of silica fume and expanded perlite addition on the technical properties of the fly ash–lime–gypsum mixture. Constr Build Mater. 22 1299 doi:10.1016/j.conbuildmat.2007.01.011

[4] Gencel O, del Coz Diaz J, Sutcu M et al. 2014 Properties of gypsum composites containing vermiculite and polypropylene fibers: Numerical and experimental results. Energy Build. 70 135 doi:10.1016/j.enbuild.2014.01.027

[5] Sayil, B. and Gürdal, E. 1999 The physical properties of polystyrene aggregated gypsum blocks. In: 8th International Conference on Durability of Building Materials and Components. Vancouver: NATIONAL RESEARCH COUNCIL CANADA, 496

[6] San-Antonio-González A, Del Río Merino M, Viñas Arrebola C, Villoria-Sáez P. 2015 Lightweight material made with gypsum and extruded polystyrene waste with enhanced thermal behaviour. Constr Build Mater. 93 57 doi:10.1016/j.conbuildmat.2015.05.040

[7] Gutiérrez-González S, Gadea J, Rodríguez A, Junco C, Calderón V. 2012 Lightweight plaster materials with enhanced thermal properties made with polyurethane foam wastes. Constr Build Mater. 28 653 doi:10.1016/j.conbuildmat.2011.10.055

[8] Serna Á, Río M, Palomo J, González M. 2012 Improvement of gypsum plaster strain capacity by the addition of rubber particles from recycled tyres. Constr Build Mater. 35 633 doi:10.1016/j.conbuildmat.2012.04.093

[9] Jiménez Rivero A, de Guzmán Báez A, García Navarro J. 2014 New composite gypsum plaster – ground waste rubber coming from pipe foam insulation. Constr Build Mater. 55 146 doi:10.1016/j.conbuildmat.2014.01.027

[10] ČSN EN 13 279-1 2014 Gypsum binders and gypsum plasters. Definitions and requirements. Czech Standardization Institute. Prague

[11] ČSN EN 196-1 Methods of testing cement – Part 1: Determination of strength 2016 Czech Standardization Institute. Prague

[12] ČSN EN 13454-2 Binders, composite binders and factory made mixtures for floor screeds based on calcium sulfate - Part 2: Test methods 2019 Czech Standardization Institute. Prague

[13] ČSN EN 933-2 Tests for geometrical properties of aggregates. Determination of particle size distribution. Test sieves, nominal size of apertures. 1997 Czech Standardization Institute. Prague

[14] ISO 468. Surface roughness. Parameters, their values and general rules for specifying requirements. 1982 International Organization for Standardization

[15] Mindess S, Young J, Darwin D 2003 Concrete (Upper Saddle River, N.J.: Prentice Hall)

[16] Krejsová J, Doleželová M, Pernicová R, Svora P, Vimmrová A 2018 The influence of different aggregates on the behavior and properties of gypsum mortars. Cem. Concr. Compos. 92 188 doi:10.1016/j.cemconcomp.2018.06.007