Optimization the composition of sand-lime products modified of diabase aggregate

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Abstract. The problem of optimizing the composition of building materials is currently of great importance due to the increasing competitiveness and technological development in the construction industry. This phenomenon also applies to catalog sand-lime. The respective arrangement of individual components or their equivalents, and linking them with the main parameters of the composition of the mixture, i.e. the lime/sand/water should lead to the intended purpose. The introduction of sand-lime diabase aggregate is concluded with a positive effect of final products. The paper presents the results of optimization with the addition of diabase aggregate. The constant value was the amount of water, variable - the mass of the dry ingredients. The program of experimental studies was taken for 6 series of silicates made in industrial conditions. Final samples were tested for mechanical and physico-chemical expanding the analysis of the mercury intrusion porosimetry, SEM and XRD. The results show that, depending on the aggregate’s contribution, exhibit differences. The sample in an amount of 10% diabase aggregate the compressive strength was higher than in the case of reference sample, while modified samples absorbed less water.

1. Introduction

The Polish construction industry is seventh in size in the European Union and it is one of the fastest growing markets in the Old Continent [1]. However, fast growing construction sector and introduction EU directives demands launching of energy-efficient and environmentally friendly solutions (harvesting raw materials, the technology of developing building materials).

Sand-lime products belong to the autoclaved materials group and therefore require a small amount of energy for hardening, they also have a natural composition so that the content of the natural background of radioactive elements in silicate products are ten times lower than the permissible use of f1 and f2 coefficient [2]. Using wall materials with high dimensional accuracy such as silicates means a reduction in the use of the chemical building by less consumption of mortar and accelerates the construction time. The increased interest and demand for sand-lime products in the Polish and Western European construction industry have an influence on the development of the scientific sphere. Currently, the composition of sand-lime products is being modified, which changes their internal structure, and consequently also the performance of the product e.g. compressive strength, water absorption, bulk density and thermal insulation.

Recently, studies were performed with liquid admixtures: lithium silicate 2.6 and 7.0, polyethylene glycol, landfill leachate as well as plastic additives and recycled materials. The application of lithium water glass (7.0) in an amount of 5% by weight of the total mixture shows that mesopores constitute
43% of all pores which is above 20% more in comparison to the reference sample. This causes reduction compressive strength from 17.29 to 5.90 MPa [3]. Introduction of lithium water glass (2.6) to traditional sand-lime products results in microstructure changes. The addition of lithium silicate in an amount of 5% decreases compression strength. Increasing the amount of lithium silicate solution nearly double the volume of mesopores (from 23% to 43%) at the expense of the macropores [4]. The use of a polyethylene glycol from 1% to 5% absorbs about 60% water less than reference sample and the use of a modifier in an amount of 1% or 5% give similar result in water adsorption test. Sample containing 1% this kind of liquid characterize improve compressive strength [5]. Conducted research allowed that the used landfill leachate contributed to achieving slightly higher compressive strength of modified sand-lime products in comparison to the traditional product (from 20.5 MPa to 21.9 MPa). More significant impact was observed in bulk density and water absorption tests (from 15.40% to 13.43%). Combination of the leachate and the water resisting admixture did not produce an expected result – higher compressive strength [6]. The addition of recycled plastics to sand-lime products can help to improve their functional properties. Suitably chosen type of polymer and its amount influences the increase in compressive strength, decrease in bulk density and water absorption as well as changes in the microstructure of resulting product. The highest average compressive strength value obtained silicates with HIPS regranulate in an amount of 20% . Samples in an amount of 30% achieves decrease water absorption of approximately 14.8% (from 16.0% to 1.2%) [7].

To the silicates, there are also added aggregates of different grain size e.g. graphite, basalt, barite. These aggregates are characterized by very high compressive strength, abrasion and other mechanical damage. However, they have to fulfill a number of standards and requirements that guarantee the longevity of formed on building materials aggregates and their safety of use. In the production of silicates, the use of barium aggregates with a grain size of 0-2 mm resulted in a significant increase in compressive strength to 44.9 MPa with a 20% aggregate contribution and to increase the bulk density of the silicate bricks [8]. Modification of silicate products with basalt aggregate resulted in a threefold increase in compressive strength, reduced water absorption from 16% to 9% relative to weight, and increased the density of the modified product [9][10].

The main raw material modifying the basic composition of sand-lime products, which is the basis of this article, is the diabase aggregate of 0-4 mm grain size. The diabase rock was extracted in the Niedźwiedzia Skala mine, located in the Małopolska province. Diabase in terms of mineral composition and origin belong to the group of gabras and basalt, where the SiO2 content is between 49.69 - 53.20 %, Al2O3 is between 15.10 – 17.50 %, FeO from 4.00 to 6.60 %, Fe2O3 and CaO is from 4.20 to 9.10% [11]. They are characterized by a density of 2.78 t/m³ [12].

The quality of the raw material used for the production of sand-lime is directly coming from the grain size and mineralogical composition which affects the physico-mechanical properties. [13] The best in quality are sands with high amount of silica, a high level of rounded grains, low content of foreign substances and good segregation of grains. The dust content should not be greater than 5%, the content of grains from 0.05 to 0.5 mm should not be less than 65 % and the minimum quartz grains should be 90 % [14]. Sand used for research is characterized by grains <0.25 mm at 5 %, 0.25-0.5 at 65% and >0.5 mm at 30 %. The observed differences in comparison with optimum are a normal phenomenon, suggesting an industrial way of extracting the raw material. Physical-mechanical parameters are not constant in sand geological layer. It is important, that the grain size curve is within acceptable tolerances, what is happening in the case with the sand from the Silicate Production Plant in Ludynia (Figure 1). The use of 0-4 mm diabase grains with a significantly predominant grain content of >0.5 mm was intended to investigate the changes in the internal structure of final products. This article aims to investigate the possibility of using the diabase aggregate as a modifier of properties of sand-lime products and to obtain answer of the question “How to use diabase aggregate additives in the sand-lime mass and get a better bricks?”.
2. Experimental

2.1. Preparation

The ready-mixed sand-lime mixture was used for experimental research and it is produced by the Silicate Production Plant in Ludynia. Samples of 40 x 40 x 160 mm were made in industrial conditions, because of it the real examine Ca/Si ratio wasn't determined. Estimated lime value in the mixture, responsible for processes that take place during the autoclave are 3% and in the planned experiment does not change. It is well known that the lime used for the production is a burnt lime and highly reactive, which is more than 90% composed of CaO, CO₂, MgO and SO₃. The dry components of the slaked lime with quartz sand of grain size distribution curve shown in figure 1 were mixed with 5% water in relation to the total weight of the product and the various contents of the diabase aggregate. The constant value was the amount of water, variable - the mass of the dry ingredients (Table 1). 6 series of 3 samples were produced. The reported results are averaged. Sample 1 is a reference sample. Sample 2 has a content of 10% diabase and sample 3 has a content of 20% diabase. Both are described in detail in the article because of demonstration the greatest differences.

Table 1. Components of sand-lime products – six samples.

| Sample  | Diabase [g] | Sand + Lime [g] | Water [g] |
|---------|-------------|-----------------|-----------|
| Sample 1| -           | 1800            | 90        |
| Sample 2| 180         | 1620            | 90        |
| Sample 3| 360         | 1440            | 90        |
| Sample 4| 540         | 1260            | 90        |
| Sample 5| 720         | 1080            | 90        |
| Sample 6| 900         | 900             | 90        |
The ingredients were stirred evenly until a damp mass was obtained, which, when kneaded in the palm, formed a compact mass. Next, the mass was placed in steel molds and compressed with a force of 25 MPa. After that, the samples were autoclaved. The temperature of hydrothermal treatment of the products is around 200°C, the pressure 1.6 MPa and the time - up to 8 hours (it means: 1h heating + 6h autoclaving + 1h cooling). The whole technological process is shown in the diagram (Figure 2).

3. Testing methods
Experimental samples were tested for physical and chemical parameters. After 21 days of the autoclaving process, the compressive strength of silicate products was tested in laboratory conditions, using a hydraulic press (Tecnotest KC 300). The samples were compressed by force proportional to the surface area of the sample.

Photos of the internal structure were made to observe hydrated calcium silicate morphology (Figure 7, Figure 8) using scanning electron microscope (SEM-type Quanta 250 FEG). Images of the scanned areas corresponding to the fragments of samples were enriched with EDS analysis. An analytical X-ray diffractometry (XRD-Empyrean, PANALYTICAL) method was used to identify the phases presented in the studied silicates. The size and number of pores in the samples were determined by mercury intrusion porosimetry (Poremaster 60, Quantachrome - USA). Porosity was calculated using the density measurements, obtained from a pycnometer.

4. Results
Testing the compression strength of silicate samples with different levels of diabase showed that it is most advantageous to use the addition of 10% of the modifier relative to the dry weight. It increases the strength from 19.2 MPa to 21.9 MPa while decreasing the water absorption (13.2%). With the significant increase in the proportion of diabase aggregates in the sample, the compressive strength of final products decreases. The sample with 20% content of the modifier shows a strength of about 14.5

Figure 2. Technological process of sand-lime products.
MPa, giving a result of over 6 MPa lower than the previous sample (Figure 3). Diabase aggregates are different from standard aggregates used to modify silicate brick, mainly mineralogical and chemical composition. Very important is the content of magnesium oxide, which is also included in the lime for the production of silicate bricks. The magnesium oxide may prevent or delay the course of an appropriate chemical reactions.

![Figure 3. Relationship between compressive strength and water absorption.](image)

The value of the porosity was obtained by multiplying the volume of mercury that entered the sample by the specific density. The sample with 10% content of diabase (specific density 2.69 g/cm$^3$) has a porosity of 39%, while a 20% diabase sample (specific density 2.62 g/cm$^3$) has 49%.

![Figure 4. Content of mesopores and macropores in the sample 2 and the sample 3.](image)
The graph (Figure 4) shows cumulative pore volume curves, depending on their diameter, obtained for the samples in question. On the x-axis (logarithmic) the pore diameters was put [μm]. On the y-axis, the volume of porous space occupied by the mercury injected into the sample was placed. The y-axis shows the percent volume of mercury. Total mercury injected into the sample gives the effective porosity of the test sample. For the sample with 10% content of diabase is 0.1454 ml/g, for the sample with 20% of diabase is 0.1865 ml/g. The graph shows that the proportion of macropores in sample 3 is 10% higher than that of sample 2 (68% - sample 2, 78% - sample 3). To sum up, increasing the amount of aggregate with a predominant proportion of 1-4 mm grain is directly proportional to the amount and pore size of the final silicate products. Assuming that these pores are unevenly distributed, we can also see that the increase in porosity with the increase in pore size is inversely proportional to the material strength.

**Figure 5.** EDS analysis of sample 2.

**Figure 6.** Microstructure of sample 2. Tobermorite.

**Figure 7.** Microstructure of sample 3.

**Figure 8.** Microstructure of sample 3.
Phase composition in sand-lime products is an important factor because it influences the performance of the product, e.g. compressive strength, porosity and absorption capacity. Crystalline phases in silicate are formed by the reaction between sand, lime and water under hydrothermal conditions, i.e. a temperature of about 200°C and a water vapor pressure of about 16 bars. Synthesis products include: C-S-H phase, most commonly found in sand-lime products, as well as tobermorite and xonotlite. It is known that 11.3 A tobermorite contains a significant amount of water, but under the influence of temperature rise (above 300 degrees) decomposes to 9A tobermorite as a result of water loss. Xonotlite is structurally similar to 11.3A tobermorite but contains 5 times less water than tobermorite and forms at ~ 220 °C - 380 °C [15].

In the case of the previously in question samples, a tobermorite phase (Figure 5, Figure 6) was formed at where also the clinotobermorite was formed (Ca₅H₂[Si₃O₉] · 5H₂O). The clinotobermorite structure is very similar to tobermorite, only a single crystals are different. [16]

The variable Ca/Si ratio has a significant effect on the formation of hydrated calcium silicates. At sample 2 with 10 % content of diabase, 92 % quartz and 3.5 % calcium carbonate were extracted (Figure 9). At sample 3 with 20 % content of diabase was observed about 6 % less quartz and about 5% more calcium carbonate (Figure 10). It is natural that the second sample has a few percent more tobermorite, while the share of the clinotobermorite decreases. In addition, diabase aggregates might be responsible for the changes in the content of individual phases in tested samples.

Figure 9. XRD patterns of sample 2.

Figure 10. XRD patterns of sample 3.
5. Conclusions
The conducted research has shown that it is most advantageous to use the addition of 10% diabase in relation to the mass of dry ingredients in sand-lime products. It has increased the compressive strength (from 19.2 to 21.9 MPa) and has reduced the water absorption of the samples at the same time (from 14.2% to 13.2%). With the significant increase in the proportion of diabase aggregates, the compressive strength of final products decreases. Increasing the content of aggregate in the samples has increased the total porosity by 10%. Moreover, the sample with 20% content of diabase has shown a greater ratio of macropores than the sample with 10% content of diabase. The use of diabase aggregates have influenced microstructural changes and the formation of the clinotobermorite phase.

Due to the above, further studies of modified sand-lime products i.e. a thorough chemical analysis, a Ca/Si ratio and studies with a smaller fraction of diabase (0-2 mm) should be carried out.

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