Mechanical properties, phase composition and microstructure of autoclaved sand – lime product

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Abstract. Autoclaved sand-lime products (silicates) are manufactured from natural raw materials and they are classified as healthy and ecological materials. A traditional raw material mix consists of quicklime (about 7% by weight), quartz sand (about 90% by weight) and water (about 3% by weight). Due to the great interest of silicate products, there have been numerous studies and modifications designed to refine the material. The article presents the research of sand – lime products. Correlations between their physical and mechanical properties and microstructure. Numerous images of scanning electron microscopy and EDS analysis contained in the article show the internal structure and phase occurring in the traditional sand - lime product and modified with different additives. The bentonite addition reduced the compressive strength by about 30% compared to the traditional sample. The addition of chalcedonite dust increased compressive strength about 20%. The microstructure has changed, both in shape, distribution and pore size, as well as the phases that have been produced in the autoclave process.

1. Introduction

Silicates are a healthy and ecological building material. Sand-lime bricks and blocks are produced exclusively from natural ingredients (90% sand, 7% lime and 3% water). Many microstructural changes take place in the process of making silicate bricks, new phases such as C-S-H and tobermory are formed [1-3]. Phase composition of building materials affects their physico-mechanical parameters such as strength, porosity and absorbency. The crystalline phases that are generated depend on the autoclaving process, the temperature, the pressure, the proportion of ingredients used, the aggregate fraction [4, 5].

Modification of the composition of sand-line products is a decisive factor in the development of this technology. The results of modification of sand-lime products aggregates with addition of basalt aggregate (2-4 mm) are known, where the addition resulted in a significant increase in the compressive strength and decrease in absorbability compared to the traditional silicate product [6]. Addition of barium sulphate BaSO₄ (0 - 2 mm) resulted in an increase in compressive strength of almost 60%, a reduction in absorbability of about 40% and a change in the microstructure of the silicate element [7, 8]. From the literature it is known to modify the composition of silicate products by the addition of metakaolinite, resulting in the formation (next to the C-S-H and tobermoryt phase) new phase – hydrogranates phase [9].

A study was conducted to determine the effect of bentonite dust and chalcedonite dust on the properties and microstructure of silicate products.
2. Materials and methods

When trying to change the quality of the sand-lime product we should focus on the favorable modification of the microstructure. To do so, we should look at the chemical reactions that occur in the product and the resulting crystalline phases.

Silicate materials are the result of the reaction between sand, lime, and water under hydrothermal conditions. This process is called autoclaving, from the name of the device in which it runs. The microstructure of silicate materials, the type of mineral phase forming, the morphology of their crystals, the volume of pores and their shape and size, are formed essentially by hardening the blocks in the autoclave.

Selecting the substrates to modify the composition of the traditional sand-lime product was guided by the knowledge of the manufacturing technology and the reactions taking place during the production process. The use of materials with high silica content is considered reasonable. Mineral dust is a waste product obtained from crushed aggregate mining, with grain size below 0.063μm. The tables below (table 1-2) show the chemical compositions of selected additives: bentonite and chalcedonite dust.

| Table 1. Chemical composition of bentonite dust. |
|-----------------------------------------------|
| SiO₂ | Al₂O₃ | CaO | Fe₂O₃ | MgO | Na₂O | H₂O | other |
| 60   | 20    | 1   | 5     | 3   | 3    | 6   | 2     |

| Table 2. Chemical composition of chalcedonite dust. |
|---------------------------------------------|
| SiO₂ | Al₂O₃ | CaO | Fe₂O₃ | MgO | Na₂O | K₂O | other |
| 90   | 5     | 0.5 | 2     | 0.25| 0.15| 0.3 | 1.8   |

The following graph (figure 1) shows the grain composition for bentonite and chalcedonite dust made using the HELOS H2746 laser diffractometer. Comparing the results of the bentonite and chalcedonite studies, we can see differences in grain size distribution. In bentonite predominate grains between 8 μm and 75 μm. Chalcedonite, on the other hand, has a regular distribution of grain size.

![Figure 1](image-url)
3. Description of research

In a silicate manufacturing plant, rectangular specimens (40x40x160 mm) were prepared. The raw material mix consists: lime (about 7% of the product weight), quartz sand (about 90%) and water (about 3%). Blocks of traditional composition and modified with additives were prepared.

Preliminary studies have been carried out to determine the effect of additives on sand-lime products. Table 3 presents the compositions of the sample series. Described as a 'N' is the traditional series, the rest containing composition modifications are described as: B – bentonite dust, C – chalcedonite dust.

| No. | Name | Content of the additive in the sample [%] | Sand-lime mass [%] |
|-----|------|------------------------------------------|-------------------|
| 1   | N    | -                                        | 100               |
| 2   | B    | 10                                       | 90                |
| 3   | C    | 10                                       | 90                |

The procedure for a mass preparation and a sample preparation was carried out in an uniform and repetitive manner. After a lime slaking process, bentonite or chalcedonite dust was added to the raw material containing sand, lime and water. After the ingredients were homogenised, the raw material was placed in moulds and pressed under 20 MPa. The samples cured in the autoclave for 8 hours at 203°C, at the pressure of 1,6 MPa. The tests were performed after the 21-day storage in dry air conditions.

Physical properties of the obtained products were determined according to the methods described in the standards for selected properties i.e. compressive strength [10], bulk density [11]. Compressive strength was determined using a Tecnotest KC 300 press. The microstructure of the products was analysed by using scanning microscopy. A Quanta 250 FEG SEM-type scanning microscope with an EDS analyzer was used.

4. Results and discussion

The results of the preliminary studies shown below (figure 2) are the averages of successive series differing in terms of applied filler. Each series consisted of 6 samples.
Figure 2 shows graph of compression strength and density. Both parameters depend on the amount and structure of pores (shape, distribution, and size). The bentonite sample has a lower bulk density and has worse compressive strength compared to the reference sample. Samples modified with bentonite dust are more porous and the strength are lower. Sand-lime products with addition of chalcedonite dust have a higher bulk density and higher compressive strength.

In the traditional way of producing silicate products, the synthesis result is usually the amorphous phase of C-S-H and crystalline tobermorite (C₅S₆H₅) [12]. There is a direct proportional relationship between the total content of these mineral phases and the strength of the autoclaved material [13]. Occurrence of other than mentioned phases can lead to different effects.

During hydrothermal treatment, C-S-H and 11.3 Å tobermorite phases are usually formed. The C-S-H phase by many researchers was referred to as tobermorite-like, currently considered to be a distinct group of varying morphology and variable phase composition. It is divided into C-S-H (I) and C-S-H (II) [14]. Tobermorite is a bonding phase, stable at a Ca / Si ratio of 0.8 to 1. Above 200°C is formed xonolite, a phase similar to tobermorite but containing less water [15, 16].

In the image of the scanning microscope for the reference sample (figure 3), the fragment of the aggregate is partially covered with the spongy C-S-H phase and the tobermorite. Tobermorite occurs in the form of sharp-pointed crystals, needles. In the fragment of the sample shown in the photo and in the other places studied, the tobermorite phase predominates. In the EDS analysis of traditional product samples (figure 4), elements such as silicon, calcium, potassium and oxygen dominate.
Modified samples also show C-S-H phases and tobermorite crystals. In samples with bentonite dust additive (figure 5), these phases occur to a much lesser extent. The EDS analysis with addition of bentonite, in addition to the silicon, calcium and oxygen elements, showed high content of aluminum and iron (figure 6). Excessive amounts of aluminum inhibit the formation of tobermorite.

In the samples with chalcedonite dust, attention is drawn to the different construction of tobermorit crystals. Compared to the sample of the traditional sand-lime product, the crystals are shorter, wider and do not have pointed ends (figure 7). In chalcedonite modified products, it can be seen that the surface being analyzed is significantly covered with a tobermorite phase. This is due to the increased strength of the samples with this additive. Studies have shown that the more fine-grained tobermorite crystals are structured, the higher the compressive strength of the material is. In addition, the EDS analysis with the addition of chalcedonite showed the dominance of the same elements as in the traditional sample, only the Si to Ca ratio was changed (figure 8).
Figure 7. Microstructure of the sample with addition of chalcedonite dust.

Figure 8. EDS analysis - sample with chalcedonite dust.

Scanning microscope observations allowed to indicate the differences in the distribution, shape and pore size of the materials tested. The pores in the sample of the traditional product (figure 9) appear at the aggregate grains contact and are irregularly shaped. Pore sizes ranging from 50 μm to 200 μm are prevalent. The pore structure in the modified samples (figure 10-11) is similar to the standard specimen (except for the bentonite additive, which has more pores with small diameters).

The size and number of pores determines the ability of the material to pull the liquid. The more capillary pores, the higher the sample will be able to draw water.

Figure 9. Pore structure of the standard sample.

Figure 10. Structure of the pores of the sample with addition of bentonite dust.

Figure 11. Structure of the pores of the sample with the addition of chalcedonite dust.

5. Conclusions

The studies showed the influence of the tested additives on the individual properties of sand-lime products. It is reasonable to continue the studies using the different percentages of the additives described to determine the optimum content of the additive and to make more conclusions about their effect on the silicate product.

Based on the research, the following conclusions can be drawn:
1. The addition of bentonite results in a reduction in the density and compressive strength of the product. In contrast, the use of chalcedonite dust increased both the bulk density and the compressive strength of the products.
2. Modifications in composition have contributed to the change in product microstructure. In samples with chalcedonite dust, more complete tobermorite crystals emerged in comparison with the traditional sample. Their content in the samples tested was very high in contrast to bentonite specimens.
3. Additives have contributed to changes in the structure and size of the pores.

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