Investigation of Physico-Chemical Properties of Sand-Lime Products Modified of Diabase Aggregate and Chalcedonite Meal

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Abstract In the era of rapid development in the construction industry, particular attention is focused on harmless and natural materials. Some of the best materials for building masonry walls are sand-lime products. Silicates are obtained from a mixture of quartz, sand and a small amount of water. They emerge as a result of the hydrothermal treatment conducted under high pressure and at a temperature of app. 203 °C. Silicates were modified of different kinds of aggregates, glass or plastics, and the content of dry ingredients was changed because of this fact. The paper describes the studies where the combination of diabase aggregate and chalcedonite meal was used. Microstructure of the products was analyzed with the use of mercury intrusion porosimetry, SEM and XRD methods. Variable content of chalcedonite meal changes the internal structure and the physico–chemical properties.

1. Introduction

Problems related to the acquisition of raw materials and fuels for the needs of the economy are more and more perceptible. Significant quantities of these resources are used in the construction and the building materials industries. In order to reduce the exploitation of non-renewable sources of natural minerals, waste products (plastics, stone meal such as chalcedonite meal) are successfully used in this branch of the economy. Fine-grained mineral additives are often used to produce the sand-lime products, so the above mentioned waste products are applied for this purpose. They form the performance of the final products. Economic and environmental aspects associated with their development are very important.

Chalcedonite is a sedimentary siliceous rock. In Poland, it occurs in the deposits located in Dębortynek, Gapinin, Lubocz and Teofilów. The only documented accumulation of this mineral in Poland is the "Teofil" deposit in the Inowłódź area. Chalcedonite is a mineral material, and its chemical and phase composition, as well as physical properties, provide the prospect of a multifaceted and multilateral use. The mineralogical composition of chalcedonite, expressed as percentage by volume, is the following: chalcedony, opal and autogenic quartz 68 - 95%, quartz and other constituents 0.5 - 6%, and free and filled pore spaces 2 - 25% [6]. Chalcedonite also has a fairly large specific surface area and a large volume of macropores. In addition, the silica that builds it is reactive [3]. Chalcedonite is characterized by high fire resistance. It should be noted that the chalcedonite deposits and the chalcedonite meal, which is obtained in the production process of the aggregate being
discussed, are characterized by a large diversity [5]. Chalcedonite meal has a bulk density of 2.63 g/cm³ [4].

According to the literature, chalcedonite meal is used in the production of concretes. The addition of chalcedonite meal (10%) has an influence on the increase of the compressive strength of several percent and on the decrease in water absorption depending on the w/c ratio and the type of cement [1].

The purpose of this article is to answer the question: Is chalcedonite meal and diabase aggregate improves the characteristics of sand-lime products? How does this combination affect the microstructure of silicate products?

2. Experimental

2.1. Preparation

40 x 40 x 160 mm sand-lime samples were used in the experiment, and they were obtained from a mixture of about 3% of lime, 5% of water and quartz sand. A lime-sand mixture prepared by Silicate Production Plant in Ludynia was used for the study. In addition, a combination of two components was used: diabase aggregate and waste product - the chalcedonite meal. Dosing of individual additives was shown in figure 1.

So far, the influence of the content of diabase aggregates on the performance of sand-lime products has been studied. It was most beneficial to use 10% of the additive in relation to the mass of dry ingredients. It has improved the compressive strength of the final products. The same 0-4 mm diabase aggregate was supplemented with dust fractions to obtain an optimal grain size distribution curve for silicates. The conducted research is an integral part of the research cycle devoted to the modification of the sand-lime products by heavy aggregates. After the first paragraph, other paragraphs are indented as you can see this in this paragraph. Please use the Vancouver numerical system where references are numbered sequentially throughout the text. The numbers occur within square brackets, like this [2], and one number can be used to designate several references. The reference list gives the references in numerical, not alphabetical, order. Please ensure that every reference cited in the text is also present in the reference list (and vice versa). Unpublished results and personal communications are not recommended in the reference list.

2.2. Testing methods

The physico-chemical properties of the experimental samples were investigated. All tests were performed under laboratory conditions. Pictures of the internal structure were made to observe hydrated calcium silicate morphology by using scanning electron microscope (SEM-type Quanta 250 FEG). Images of the scanned areas corresponding to the fragments of the samples were enriched with EDS analysis. An analytical X-ray diffractometry (XRD-Empyrean, PANALYTICAL) method was used to identify the phases occurring in the studied silicates. The size and the 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.
Considering the physical characteristics, humidity, density and compressive strength were examined. After 21 days of the autoclaving process, the compressive strength of the silicate products was tested in laboratory conditions using a hydraulic press (Tecnotest KC300), according to the methodology given in the standard.

3. Results and discussion

![Sample A Microstructure](image1)

**Figure 2.** Microstructure of sample A

![XRD Analysis](image2)

**Figure 3.** XRD Analysis of sample A.

Crystalline phases in silicate are developed as a result of the reaction between CaO-SiO$_2$-H$_2$O under hydrothermal conditions, i.e. a temperature of about 180-200°C and a water vapor pressure of about 16 bars [7]. Synthesis products include: C-S-H phase, most commonly found in sand-lime products, as well as tobermorite and xonotlite. CSH phase is clearly amorphous (sometimes spongy) or takes the fibrous form. Tobermorite (Ca$_5$Si$_6$O$_{17}$*5H$_2$O) is characterized by the regular lamellar shape.
High strength phase formed during autocalving is present in the form of long fibers with sharp endings, visually resembling needles [8].

The XRD analysis of the examined samples has shown the existence of minerals typical for silicates in the microstructure, i.e. quartz, which is composed mainly of silicon dioxide, and it is a basic component of silicates, aragonite and calcite (multiform varieties of calcium carbonate CaCO₃ and products of hydrothermal processes) [9].

The basic element of the structure of the tobermorite of hydrated calcium silicates is cation sublattice consisting of octahedra. On this sublattice, silicon-oxygen tetrahedra condense, and in the case of CSH phase, they form very disordered elements of triclinic wollastonite chain, the consequence of which is an alternating relation of Ca/Si, and in the case of tobermorite 11.3, a correct structure of the wollastonite chains occurs. Transferring of one phase into the other requires supplementing the absent segments of wollastonite chain and the spatial extension of crystal structures. In sample A (figure 2 and 3), the occurrence of tobermorite in the amount of 2.2% was proved, and the occurrence of clinotobermorite in the amount of 0.8% was proved as well, and it is
structurally similar to the tobermorite. The literature says that clinotobermorite occurs as tabular crystals parallel to the c-face with up to 5 mm width and the structures of clinotobermorite and tobermorite are very similar.

Figure 8. Content of mesopores and macropores in the sample A and in the sample B

Figure 9. Results of compressive strength

Figure 8 shows the cumulative curves of the volume of the pores, depending on their diameter, obtained for sample A and sample B. The x-axis includes the diameters of the pores, in the range available for porosimeter, from 001 to 100 µm. The y-axis shows cumulative volume of porous space
occupied by the mercury intruded to the sample [%]. The total volume of mercury intruded to the sample gives the result of effective porosity. The curves for sample A and sample B show comparable amount of mesopores. The biggest differences are visible in the amounts of pores of bigger diameters. It is inferred that the cause may be the increase of chalcedonite meal from 10 to 20%. The curves for the studied samples are shaped in such a way that it is indicative of the presence of the porous space deviating from the cylindrical model.

Table 1. Results of volume intruded, specific density and porosity in sample A and B

|          | Volume intruded [cm³/g] | Specific density [g/cm³] | Porosity [%] |
|----------|-------------------------|--------------------------|--------------|
| Sample A | 0.2012                  | 2.6182                   | 52.68        |
| Sample B | 0.2078                  | 2.5822                   | 53.56        |

Table 1 shows the results of the measurements carried out with the use of mercury intrusion porosimeter and the total porosity, calculated with the use of the results of volume measurements. Samples A and B show porosity on a similar level, from 52% to 53.5%. It is an unusually high value, because the reference sample has porosity of about 37%. The studies that have been carried out so far show that the samples with 10% of diabase aggregate achieve porosity on the level of about 39%. It is therefore inferred that the addition of chalcedonite meal has an influence on the significant increase (of about 13-14%) of the effective porosity.

4. Conclusions

It results from the micro- and macroscopic observations that the content of an increased amount of chalcedonite meal in lime-sand products has an influence on the disappearance of tobermorite phase. Its absence results in the mechanical properties of the silicates.

The combination of 10% of diabase aggregate and 10% of chalcedonite meal shows the best mechanical properties. It is therefore observed that the increase of compressive strength is of about 4% in relation to the reference sample, while the humidity is maintained on the level of 15%.

In the case of constant amount of diabase aggregate in the sample, regardless of the amount of chalcedonite meal, the samples show a comparable effective porosity - about 52-53.5%. The samples show the differences in terms of the macropores amount distribution.

5. References

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