

Autoclaved materials with chalcedonite addition

Ryszard Dachowski*, Paulina Kostrzewa , and Sylwia Brelak

Kielce University of Technology, *Faculty of Civil Engineering and Architecture*, Kielce, Poland

Abstract. Silicate products along with autoclaved aerated concrete are a healthy and ecological building material. Silicates and AAC are the effect of the reaction of the components from which they are formed under hydrothermal conditions. This process is called autoclaving from the name of the devices in which it runs. The tests have shown the effect of the tested additives on the particular properties of autoclaved lime-sand products and lime-sand-cement products. The addition of chalcedonite powder increased both the bulk density and compressive strength of lime-sand products and lime-sand-cement products. Composition modifications contributed to changing the microstructure of the products. In the samples with the addition of chalcedonite powder, fuller tobermorite crystals developed in comparison with the traditional sample. Additives contributed to changes in pore structure and size.

1 Introduction

The sand-lime brick, commonly known as silicate, has been produced on a massive scale for nearly 130 years. Only sand, lime and water are used for its production. It makes it hard to find a material more natural than silicate. Easy access to raw materials, their naturalness and relatively cheap production process caused that silicates belong to one of the most popular materials used in housing construction. [1-3]. Another popular wall building material is autoclaved aerated concrete (AAC). Due to the lack of hazardous substances, like silicate is a material that creates a healthy environment for human life and does not adversely affect the natural environment.

Modification of the composition of construction products is their decisive development factor. Autoclaved products have been subjected to modifications of composition and production technology many times. There are known, for example, the results of tests of lime-sand products with the addition of basalt aggregate (2 - 4 mm) [4], BaSO₄ barium sulphate (0 - 2 mm) [5, 6], metakaolinite [7], bentonite [8]. The literature gives modifications of autoclaved aerated concrete using halloysite [9] or plastics [10, 11].

In the study, research was undertaken to determine the effect of the addition of chalcedonite powder on the properties and microstructure of autoclaved lime-sand and limestone-sand-cement products.

* Corresponding author: pkostrzewa@tu.kielce.pl

2 Materials and methods

When undertaking an attempt to change the quality of a construction product, it is necessary to focus on the favorable modification of the microstructure. To do this, look at the chemical reactions in the product and the emerging crystalline phases.

Silicates and AAC are the effect of the reaction between the components under the hydrothermal conditions from which they are produced. This process is called autoclaving from the name of the devices in which it runs. The microstructure of silicate blocks and AAC, the type of mineral phases formed, their crystal morphology, the volume proportion of pores and their shape and size, are shaped basically during processing of products in an autoclave.

When selecting substrates for the modification of the traditional composition of sand-lime and limestone-sand-cement products, knowledge of manufacturing technology and reactions taking place during the production process was followed. The use of high silica material was considered justified. The table below (Table 1) presents the chemical composition of the selected additive: chalcedonite.

Table 1. Chemical composition of ground chalcedonite.

SiO ₂	Al ₂ O ₃	CaO	Fe ₂ O ₃	MgO	Na ₂ O	K ₂ O	other
90	5	0,5	2	0,25	0,15	0,3	1,8

The following graph (figure 1) shows the grain size determination for chalcedonite powder made using the HELOS H2746 laser diffractometer. Chalcedonite has an even distribution of grain size. The distribution of grain diameters is in the range between 0.3 μm and 90 μm.

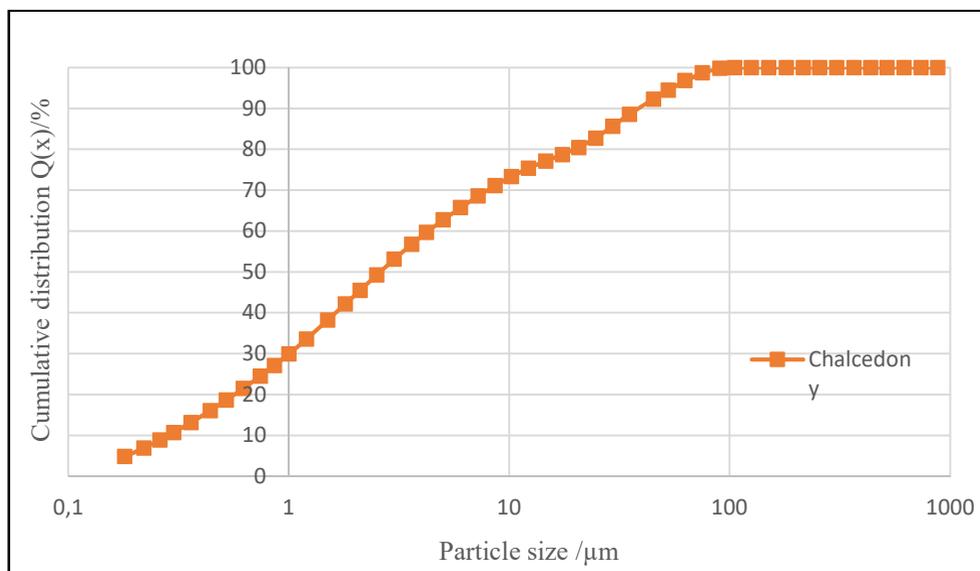


Fig. 1. The granulometric composition of chalcedonite.

3 Tests description

For the silicate brick, rectangular samples (size 40x40x160 mm) were prepared. The raw mix consists of: quick lime (about 7% of the product's weight), quartz sand (about 90%) and water (about 3%). Autoclave aerated concrete has been prepared with cubic samples (100 mm side). The raw material mixture included: sand combined with gypsum (about 72% of the product weight), cement and lime (about 20%) and water (about 7%), as well as aluminum paste and additives improving the rheological properties of the mass. In both cases, blocks with a traditional composition and modified additives were prepared.

In the first phase of the production process of lime-sand products, the sand was mixed with burnt lime and water in appropriate proportions and placed in reactors, where the process of lime mass extinguishing took place. Subsequently, the substrate was added in a percentage to the weight of the raw material. After mixing all ingredients and moisturizing the silicate mass, it was placed in molds and pressed at 20 MPa. The samples were then left in autoclaves for 8 hours at 203°C and 1.6 MPa. In this way, the prepared samples after demoulding were stored for 21 days under appropriate temperature and humidity conditions.

The raw material mixture for autoleveled aerated concrete was prepared in semi-industrial conditions in SW technology with the version 500. In this technology, sand sludge is made by grinding sand with water. The components in the right order are transferred to the stationary mixer. In a weighing way, the mixer is filled with a weighted portion of sand sludge and water, and the surfactant is dosed. Mixing takes place for about 1-2 minutes. Then the binder is added and the mixture is stirred again for about 2.5-3 minutes. In the next stage, aluminum powder is added in the aqueous solution of the surfactant and mixing for approx. 1.5-2 min. At this stage, appropriate amounts of additives have been added to the profit. After mixing, the product was poured into three-part forms, and these were placed in pre-ripening chambers where at about 60°C they remained for about 3 hours. After demoulding, the samples underwent autoclaving for approx. 13 h of hydrothermal treatment at 191°C and pressure water vapor 1.2 MPa.

Tests of physical properties of the obtained plastics were carried out in accordance with the scope and methods given in standards relating to selected features: compressive strength [12], dry density in the dry state [13]. Compressive strength was determined using the Tecnotest KC 300 press. The microstructure of materials obtained under hydrothermal conditions was examined by scanning microscopy. SEM-type Quanta 250 FEG with the EDS analyzer was used in the conducted research.

4 Results and tests analysis

The results of the preliminary tests presented below (figure 2) are average values of subsequent series differing in terms of the filler used. Six samples consisted of each series. The 'S' series is marked by a series of autoclaved lime-sand products and 'A' series of autoclaved limestone-sand-cement products. Ch10 means the addition of 10% chalcodinite powder in relation to the sand-lime mass, and likewise Ch20 means 20% addition.

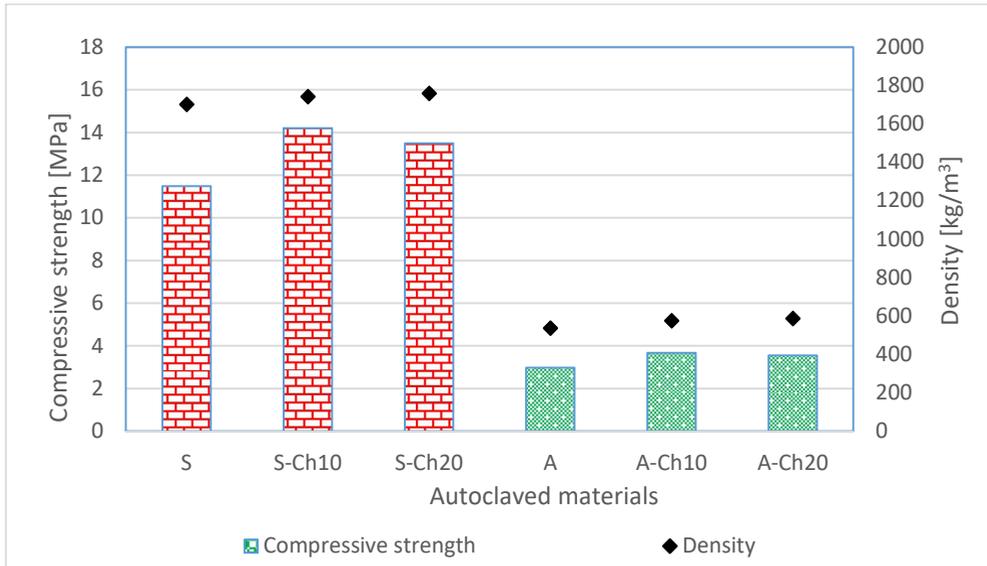


Fig. 2. Compilation of compressive strength and bulk density of traditional and modified products.

Figure 2 shows a graph of compressive strength and density. Both parameters depend on the number and structure of pores (shape, distribution and size) in the material. Silicates have even four times higher density than autoclaved aerated concrete, which also translates into compressive strength. Silicate on average achieves compressive strength between 10-15 MPa and AAC approximately 4 MPa. Chalcedonite powder did not significantly contribute to the change in the bulk density of products, however, as compared to standard samples, an increase in compressive strength is visible.

In the traditional production of silicate products, the synthesis products are usually the amorphous C-S-H phase and the crystalline tobermorite ($C_5S_6H_5$) [14]. Tobermorite is a binding phase, stable in Ca/Si ratio from 0.8 to 1. Above 200°C, xonotlit, a tobermorite-like phase but containing less water [14-16] is formed.

In traditional products from autoclaved aerated concrete, basic phase products include: tobermorite ($C_5S_6H_5$) and C-S-H phase in submicrystalline form [14]. In smaller amounts, there is also anhydrite (C) and hydrogranates ($C_xA_yS_mH_n$), as well as in small amounts xonotlit ($C_6Si_6H_2$) and pectolite (NC_4S_6H) [17-19].

For both silicates and autoclaved aerated concrete, there is a direct relationship between the total content of these mineral phases and the strength of the autoclaved material. The occurrence of others in addition to the mentioned phases that bind individual grains of sand may lead to different effects. During hydrothermal treatment, the C-S-H and 11.3 Å tobermorite phases usually occur. The C-S-H phase, by many researchers, was termed as a tobermorite-like, now it is considered to be a separate group with different morphology and variable phase composition. Taylor [19] divides her into C-S-H (I) and C-S-H (II). According to Diamond [20], there are four morphological types of the C-S-H phase. It distinguishes C-S-H I on fibrous structure, C-S-H II so-called "Honeycomb" in the form of a grid corresponding to the C-S-H (I) phase of Taylor, C-S-H III with isometric grains structure and C-S-H IV forming spherical assemblies. The occurrence of individual phases influences the strength of autoclaved products. The C-S-H phase improves the increase in strength, while its reduction is caused by a large amount of calcium and a low content of active silica, eg when α C₂SH is formed [17].

In the photo from the scanning microscope reference sample of the silicate (figure 3) is visible fragment of the aggregate partially covered with spongy phase C-S-H and tobermorite.

Tobermorite occurs in the form of pointed crystals and needles. In the fragment of the sample shown in the picture as well as in other places studied the participation of the tobermorite phase prevails. In the EDS analysis (figure 5), elements such as silicon, calcium, potassium and oxygen predominate.

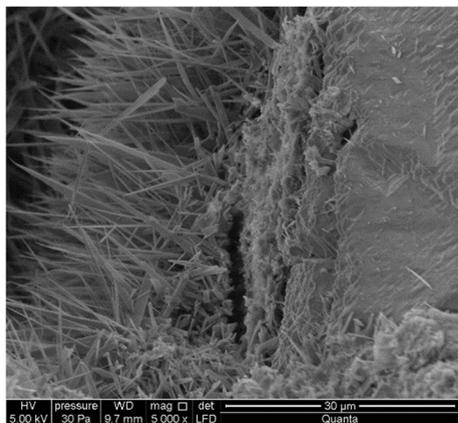


Figure 3. Microstructure of a standard silicate sample.

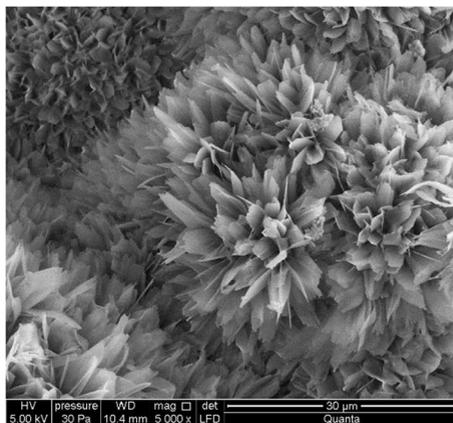


Figure 4. Microstructure of a silicate sample with the addition of chalcedonite powder.

In the modified samples (Figure 4), the C-S-H phase is also observed at the interface with the aggregate and tobermorite crystals. In samples with chalcedonite powder attention is drawn to the different structure of tampermorite crystals. In comparison with a sample of a traditional sand-lime product, the crystals are shorter, wider and have no pointed ends. In the studied fragments of samples modified with the addition of chalcedonite powder, it can be noticed that the analyzed surface is significantly covered with the tobermorite phase. This is due to the increased strength of the samples with this addition. Research has shown that the more tiny tobermorite crystals with a regular structure, the greater the compressive strength of the material. In addition, the EDS analysis of the silicate sample with the addition of chalcedonite powder showed the dominance of the same elements as in the case of a traditional silicate sample, only the ratio of Si to Ca (Figure 6) changed.

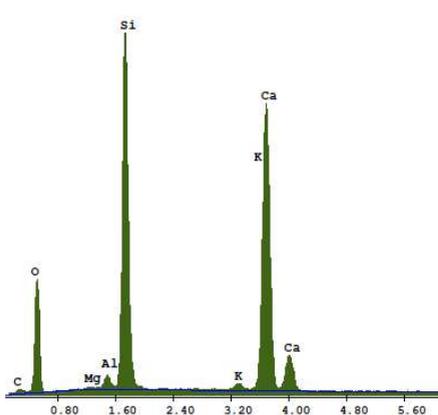


Figure 5. EDS analysis of a standard silicate sample.

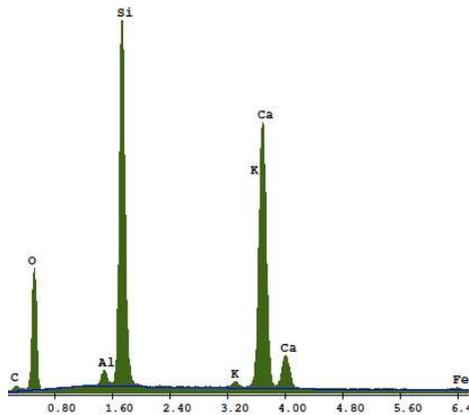


Figure 6. EDS analysis of a silicate sample with the addition of chalcedonite powder.

The microstructure of the retort sample of autoclaved aerated concrete is shown in Figure 7. The tobermorite and C-S-H phases were observed in the test sample. Tobermorite occurs in the form of pointed needles, whereas C-S-H in fibrous form. EDS analysis of this sample (figure 9) showed the dominance of the elements Si, Ca, K, O and in a smaller amount of Al.

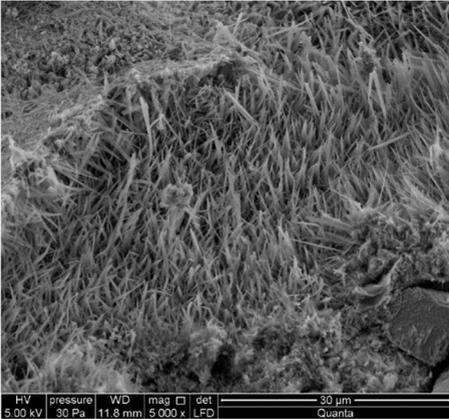


Figure 7. Microstructure of the AAC standard sample.

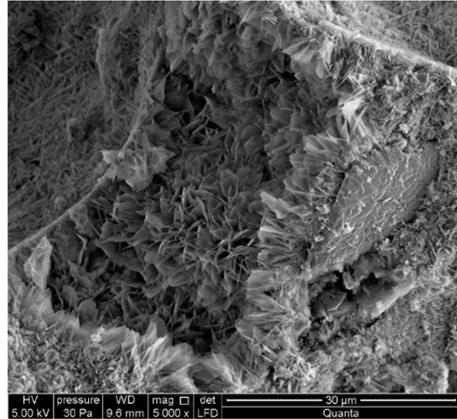


Figure 8. Microstructure of the AAC sample with the addition of chalcidinite powder.

Figure 8 presents the microstructure of a sample of modified autoclaved aerated concrete made by scanning microscope. Similarly to the traditional ACC sample, numerous C-S-H and tobermorite phases were also observed here. Numerous clusters of crystals with a lamellar habit have been observed, these crystals are wide and have an oval end. The EDS analysis of AAC samples with the addition of chalcidinite powder (figure 10) showed the dominance of the same elements as in the case of the traditional ACC sample, but O and Al are present in a smaller amount than in the traditional sample.

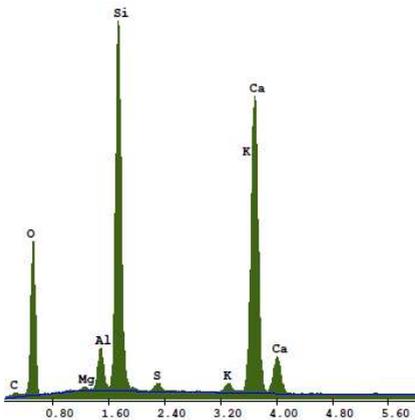


Figure 9. EDS analysis of the AAC standard sample.

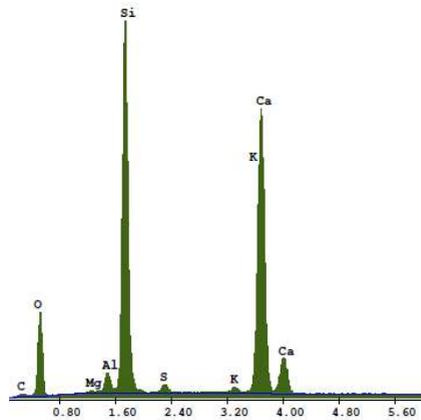


Figure 10. EDS analysis of the AAC sample with the addition of chalcidinite powder.

5 Summary and conclusions

The tests have shown the effect of the tested additives on the particular properties of autoclaved lime-sand products and lime-sand-cement products. It is reasonable to continue research using different percentages of the additives described, in order to determine the optimum content of the additive and more complete conclusions regarding their impact on the mentioned products.

The following conclusions can be drawn on the basis of the research:

1. The addition of chalcidonite powder increased both the bulk density and compressive strength of lime-sand products and lime-sand-cement products.
2. Composition modifications contributed to changing the microstructure of the products. In the samples with the addition of chalcidonite powder, fuller tobermorite crystals developed in comparison with the traditional sample.
3. Additives contributed to changes in pore structure and size.

References

1. Z. Pytel, Modification of the phase composition and microstructure of autoclaved sand-lime bricks, *Ceramics*, **116** (2014)
2. S. Wolfke, *Technology of Sand-Lime Products*, Arkady, (1986)
3. A. Stępień, The Impact of Glass Additives on the Functional and Microstructural Properties of Sand-Lime Bricks, *International Journal of Civil and Environmental Engineering* **4**, 3 (2017)
4. A. Stępień, Microstructural and functional properties of silicate products modified with basalt aggregate, *Structure and Environment*, **1** (2012)
5. A. Stępień, The Impact of Barium Sulfate on the Microstructural and Mechanical Properties of Autoclaved Silicate Products, *Technika* (2014)
6. R. Dachowski, S. Stępień, The Impact of Barium Aggregate on the Microstructure of Sand-Lime Products, *Trans Tech Publications LTD* (2011)
7. Z. Pytel, Investigations on the Influence of Hydrogarnets Presence on the Properties of Sand-Lime Bricks (2011)
8. Z. Owsiak, P. Kostrzewa, *IOP Conf. Ser.: Mater. Sci. Eng.* **251** (2017)
9. Z. Owsiak, A. Sołtys, The influence of a halloysite additive on the performance of autoclaved aerated concrete *Ceramics-Silikaty* **59**, 1 (2015)
10. R. Dachowski, S. Kapała, Modification of autoclaved aerated concrete with high-impact polystyrene *Materiały budowlane*, **6** (2016)
11. S. Brełak, R. Dachowski, Effect of Autoclaved Aerated Concrete Modification with High-Impact Polystyrene on Sound Insulation *WMCAUS IOP Publishing IOP Conf. Series: Materials Science and Engineering* **245** (2017)
12. PN-EN 772-1+A1:2015-10E Methods of test for masonry units - Part 1: Determination of compressive strength
13. PN-EN 772-13:2001P Methods of masonry units – Part 13: Determination of net and gross dry density of masonry units (except for natural stone)
14. G. Zapotoczna-Sytek, S. Balkovic, *Autoclaved Aerated Concrete*, PWN (2013)
15. A. Hartmann, J. C. Buhl, K. Breugel, Structure and phase investigations on crystallization of 11 Å tobermorite in lime sand pellets, *Cement Concrete Res.* **37** (2007)
16. S. Shaw, S. M. Clark, C. M. B. Henderson, Hydrothermal Formation of the Calcium Silicate Hydrates, Tobermorite ($\text{Ca}_5\text{Si}_6\text{O}_{16}(\text{OH})_2 \cdot 4\text{H}_2\text{O}$) and Xonotlite ($\text{Ca}_6\text{Si}_6\text{O}_{17}(\text{OH})_2$): an in Situ Synchrotron Study, *Chemical Geology*, **167** (2000)
17. P. Gębarowski, K. Łaskawiec, M. Skoroniewska, Wpływ warunków autoklawizacji na

- właściwości tworzyw krzemianowych, Prace ICiMB **20** (2015)
18. W. Kurdowski, Chemia cementu i betonu, Wydawnictwo Polski Cement Wydawnictwo Naukowe PWN (2010)
 19. H. F. W. Taylor, Cement Chemistry, Academic Press (1990)
 20. S. Diamond, Material Research and Society Symposium Proceedints, **2** (1981)