THE INFLUENCE OF A HALLOYSITE ADDITIVE ON THE PERFORMANCE OF AUTOCLAVED AERATED CONCRETE

Z. OWSIAK, #A. SOŁTYS

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

#E-mail: soltys_a@wp.pl

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This paper presents the results from the tests of autoclaved aerated concrete with halloysite as a cement additive. Good pozzolanic properties make it a suitable material to be used as a partial replacement of a portion of cement. Basic physical and mechanical properties of the composites with various mineral content are discussed. The compressive strength test results indicate an increase in strength of the AAC containing 2.5 % and 5 % halloysite relative to the reference specimen. Thermal conductivity and density values remained at the same level. Observations of the microstructure in the scanning electron microscope confirmed the results from the XRD tests. Anhydrite was observed in addition to tobermorite. The results from the tests of the autoclaved aerated concretes in which halloysite was incorporated as 7.5 % and 10 % cement replacement showed an increase in compressive strength, density and thermal conductivity values.

INTRODUCTION

Autoclaved aerated concrete (AAC) is primarily made with sand and a mixture of cement, lime and gypsum as a binding agent. Use of mineral additives in aerated concrete formulations is increasing, as is their importance [1, 2]. One of the minerals that can be added to the hydraulic binder is halloysite, a mineral from a kaolinite group with the chemical formula $Al_4[Si_4O_{10}](OH)_8{\cdot}4H_2O$. Halloysite is a 1:1 two-layered aluminosilicate composed of sheets shifted relative to each other in directions of *a-axis* and *b-axis*, with a highly disordered layer sequence and stratification. The tubular morphology of the mineral is a result of the relief of structural strains that arise from a misfit between tetrahedral and octahedral sheets. Water particles present between the sheets form a monomolecular layer and contribute to the weakening of hydrogen bonds [3, 4].

The chemical composition and physical properties of halloysite make it suitable for improving the characteristics of cement composites. In the pore solution of the cement paste, halloysite is subject to dissociation and the resultant aluminate ions may form calcium aluminosilicates. The silica present on the halloysite surface may react with calcium ions and form calcium silicate hydrate [5].

The aim of this study was to assess the effect of the halloysite additive on the properties of the slow-setting silicate-based autoclaved aerated concrete (SW technology). The values of water absorption, thermal conductivity, density and compressive strength were determined through physical and mechanical testing. In addition, the testing programme included evaluation of the influence of the halloysite in the amount from 0 to 10 % of the cement mass on the phase composition and the microstructure of the autoclaved aerated concrete.

MATERIALS AND METHODS

The autoclaved aerated concrete was prepared from cement CEM II/A-V, lime, sand, gypsum and halloysite from the Dunino deposit in accordance with the requirements of the relevant standards. The w/c ratio was 0.47 (w/c = 0.47). The halloysite was added in the amount of 2.5; 5.0; 7.5 and 10 % by mass of cement. The aggregate/binder ratio was 2.54. Table 1 shows the chemical composition of the halloysite; its microstructure (SEM) and X-ray analysis (EDS) are shown in Figure 1.

The microstructure in Figure 1 reveals that the mineral is made of dispersed nanotubes and nanolaths. An X-ray analysis of the microregion confirmed the

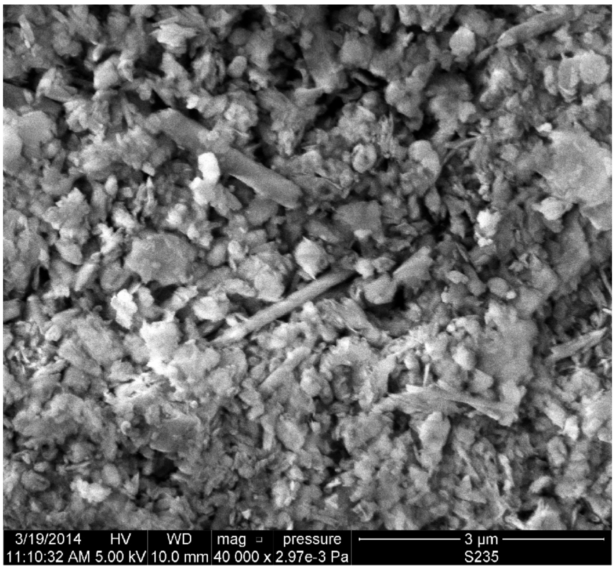
Table 1. Oxide composition of halloysite.

Oxides	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	P_2O_5	CaO	Na_2O	MgO	K_2O	MnO
Content (%)	49.6	41.54	5.66	1.98	0.79	0.25	0.08	0.063	0.063	0.046

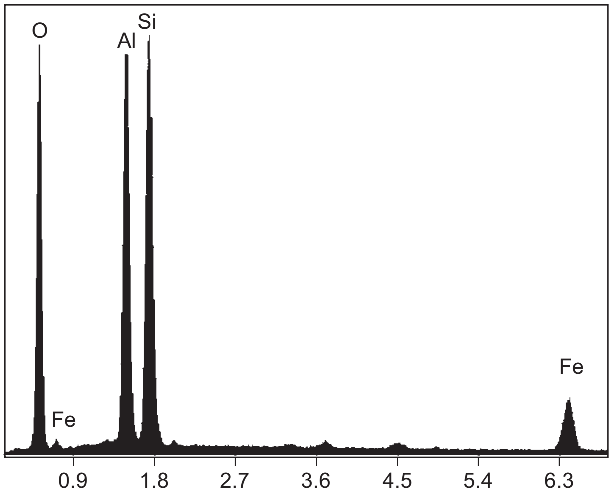
results from the chemical composition tests and indicated silicon, aluminium and iron as its main constituents.

The designation of the mixtures with a varied amount of halloysite is shown in Table 2.

The output components in suitable proportions were homogenized for 5 minutes in the stationary mixer. The mixture was then cast in Æ8 cm moulds. The specimens, kept in the moulds, were stored in the curing chamber for 5 hours at a temperature of 60°C and humidity exceeding 90 %. After that time the specimens were demoulded and placed in an autoclave for 8 hours at a temperature of 186°C.



a)



b)

Figure 1. Micro-structure (SEM) and microanalysis (EDS) of halloysite.

Table 2. Designation of AAC specimens.

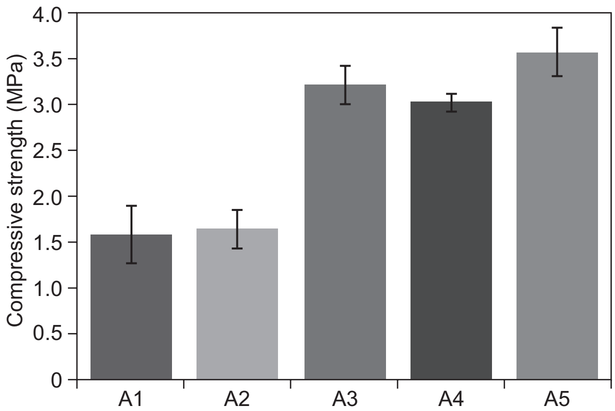
SPECIMEN	A1	A2	A3	A4	A5
Halloysite content (%)	0	2.5	5	7.5	10

Prior to physical and mechanical tests, the specimens were dried to the constant mass at a temperature of 50°C. The compressive strength test was carried out in accordance with PN-EN 771-1, the density was determined to PN-EN 772-13, the heat transfer coefficient was measured in accordance with PN ISO 8301, and the capillary absorption was determined following the procedures laid out in PN-EN 772-11:2002. The phase composition of the composites was analysed using the X-ray EMPYREAN diffractometer by PANalytical. Observations of the microstructure were conducted in the scanning microscope Quanta 250 FEG at high vacuum.

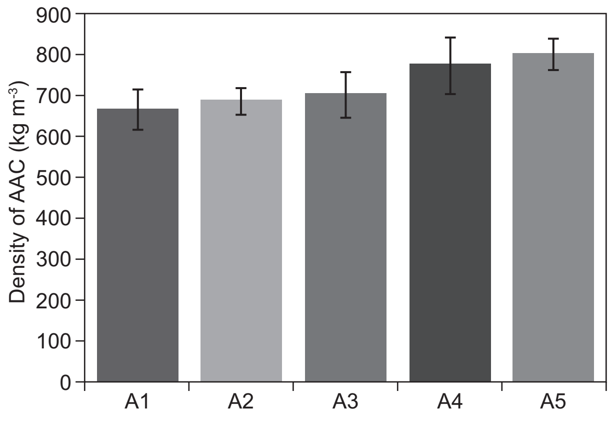
RESULTS

Figures 2a and 2b show the results from the compressive strength and density tests. The value of the compressive strength of the autoclaved aerated concrete with an addition of 2.5 % halloysite (A2) was 4 % higher than that of the reference specimen (A1). The 5 % replacement (specimen A3) led to a two-fold increase in strength.

Analysis of the density (Figure 2b) of the autoclaved aerated concretes indicates that density values of A1, A2



a)



b)

Figure 2. Compressive strength (a) and density (b) of AAC (A1 – 0 %; A2 – 2.5 %; A3 – 5.0 %; A4 – 7.5 %; A5 – 10 %).

and A3 were within the range 670 - 700 kg·m^{-3}. Mixtures A4 and A5 had the density of 772 and 798 kg·m^{-3}. The incorporation of 7.5 and 10 % mass fraction of halloysite contributed to the increase in density and, as a result, to the increase in the overall strength of the concrete.

Thermal conductivity values of autoclaved aerated concretes with various content of halloysite are shown in Figure 3.

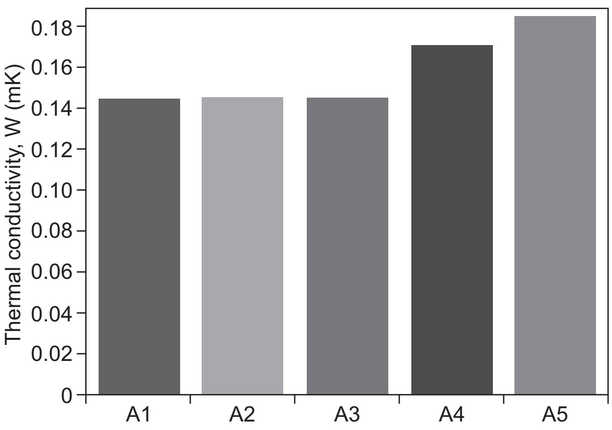


Figure 3. Thermal conductivity of AAC with halloysite additive (A1 – 0 %; A2 – 2.5 %; A3 – 5.0 %; A4 – 7.5 %; A5 – 10 %).

Halloysite used as a substitute for 2.5 % and 5 % of cement raised the thermal conductivity of the AAC up to 0.146 W/(m·K), which was the level presented by the AAC without the halloysite. Higher amounts of halloysite added, i.e., 7.5 % and 10 %, provided thermal conductivity increase of 17 % and 28 %, respectively.

Table 3 shows the results from the capillary absorption tests.

Table 3. Capillary absorption of water in AAC.

Test duration (min)	Specimens				
	A1	A2	A3	A4	A5
	Capillary absorption of water (g m^{-2} s$^{-0.5}$)				
10	191	204	221	193	180
30	93	95	97	94	89
90	42	44	43	43	41

The results from the tests for the capillary water absorption by the surfaces of the autoclaved aerated concrete specimens after 10 minutes presented some differences. Compared with the reference specimen, the lowest value of the capillary absorption was observed for specimen A5, whereas the highest for specimen A3. After 30 and 90 minutes, all the specimens tested had similar capillary absorption values. Capillary water absorption was found to be the function of the property of the material. This property is particularly important when aerated concrete is to be used in the outer elements of a building as a finish, and its value varies across products and manufacturers [7].

Figure 4 shows the diffractograms of the autoclaved aerated concretes with various halloysite content.

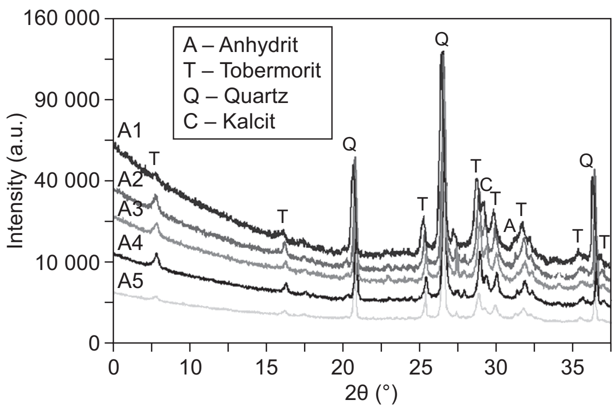
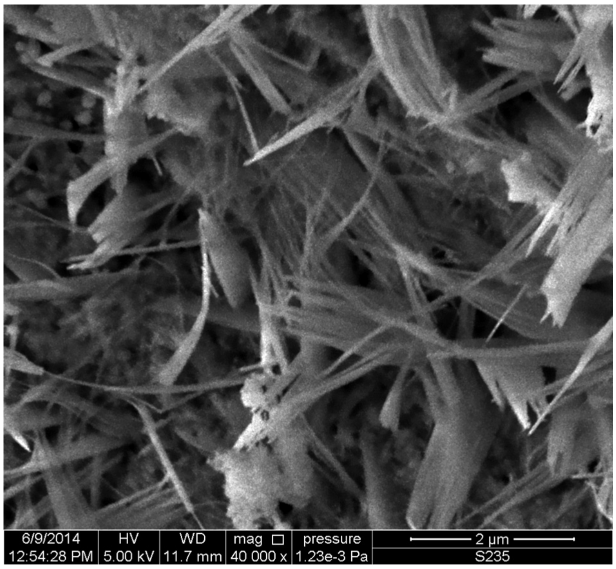


Figure 4. X-ray diffraction patterns of AAC (A1 – 0 %; A2 – 2.5 %; A3 – 5.0 %; A4 – 7.5 %; A5 – 10 %).

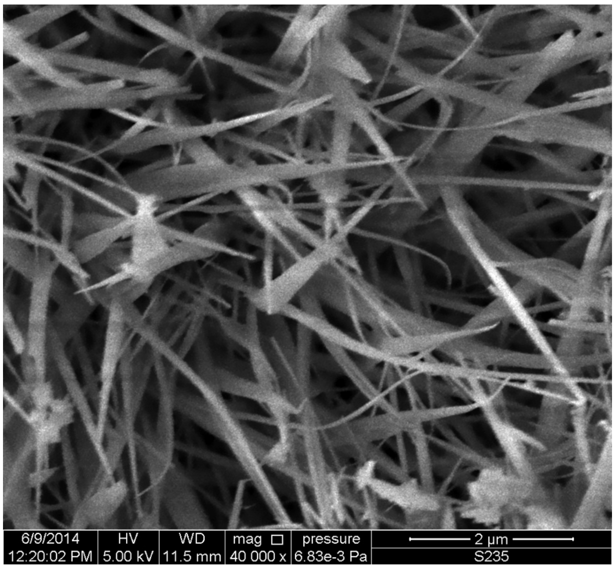
The X-ray analysis of the concrete specimens with halloysite revealed main reflections typical of quartz and tobermorite, and peaks of low intensity from anhydrite and calcite. Compared with those in the 2.5 and 5 % halloysite specimens, tobermorite peaks in the reference sample had a lower intensity. The intensity of the calcite peak decreased with the increase in the quantity of the mineral from the kaolinite group. The presence of this phase is attributed to the carbonation of unreacted lime. The peaks of quartz are related to the increased quantity of SiO_2 introduced with the halloysite.

Figure 5 shows the microstructure of the autoclaved aerated concrete observed in the scanning microscope.

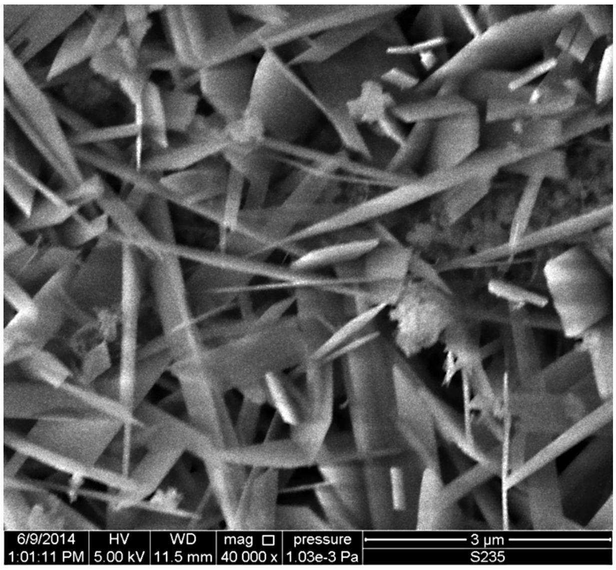


a) A1

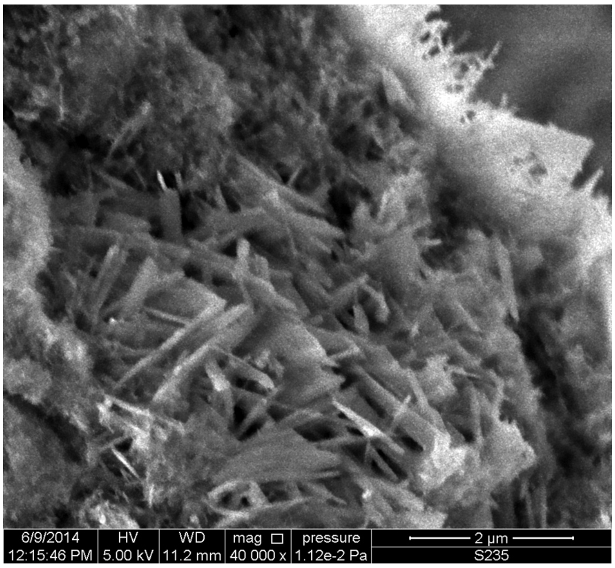
Figure 5. AAC microstructure.



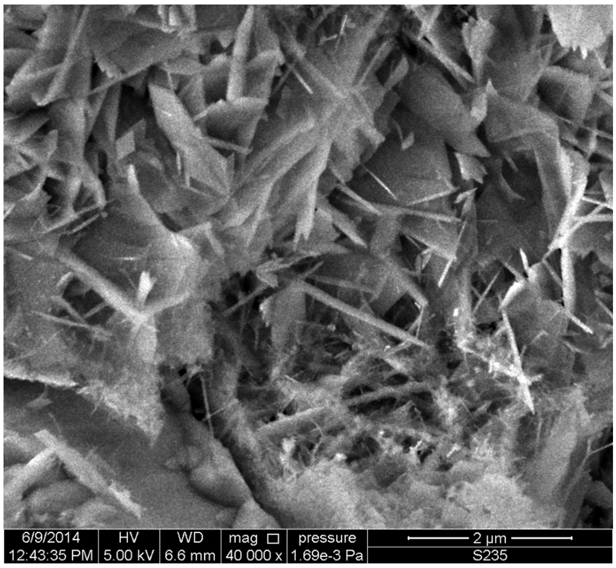
b) A2



c) A3



d) A4



e) A5

Figure 5. AAC microstructure (*continue*).

Agglomerations of tobermorite are seen in the specimens denoted as A1, A2 and A3. Anhydrite was detected in the aerated concrete with 7.5 and 10 % halloysite replacement of cement.

DISCUSSION AND SUMMARY

The results from the study lend strong support to the conclusion that halloysite used as a cement mineral additive plays a significant role in the ultimate performance of the autoclaved aerated concrete. The results described in this paper indicate the increase in the compressive strength of the AAC with 2.5 % and 5 % halloysite content, compared with the reference specimen at the similar values of density. The analysis of the phase composition, conducted to explain the differences in the compressive strengths of the specimens, showed the increase in the intensity of peaks derived from tobermorite for A2 and A3. Two mechanisms are responsible for the increase in strength. The first mechanism is associated with the fact that halloysite may act as a filler due to its nano-sized particles. The mineral may fill the voids formed after the evaporation of water from the autoclaved composites. The other mechanism relates to a high content of SiO_2 and hydroxyl groups present on the halloysite surface. The hydroxyl groups may react with Ca^{2+} ions from the cement paste resulting in the higher content of C–S–H phase, which under hydrothermal conditions and with progressing synthesis, is transitioning into tobermorite1.1nm [5, 7]. Neither 2.5 % nor 5 % content of halloysite affects the thermal conductivity change, compared with the reference specimen. The observations of the microstructure in the scanning microscope confirmed the XRD results. In addition to tobermorite, anhydryte was observed. The results for the aerated concrete in which 7.5 % and 10 % cement was replaced with halloysite showed an increase in the compressive strength, density and thermal conductivity. The tests conducted to determine physical and mechanical properties of autoclaved concrete with various halloysite contents showed the best results for the 5 % addition of halloysite by mass of cement. The density was similar to that presented by the concrete without the additive. A two-fold increase in compressive strength was obtained. In the autoclaved aerated concrete with an addition of 2.5 and 5 % of halloysite, the correlation between the compressive strength and the density of the material was maintained.

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