Study of Metakaolin-Based Geopolymer with Addition of Polymer Admixtures

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Abstract—In the present work, metakaolin-based geopolymer including different polymer admixtures was studied. Different types of commercial polymer admixtures VINNAPAS® and polyethylene glycol of different relative molecular weight were used as polymer admixtures. The main objective of this work is to investigate the influence of different types of admixtures on the properties of metakaolin-based geopolymer mortars considering their different dosage. Mechanical properties, such as flexural and compressive strength were experimentally determined. Also, study of the microstructure of selected specimens by using a scanning electron microscope was performed. The results showed that the specimen with addition of 1.5% of VINNAPAS® 7016 F and 10% of polyethylene glycol 400 achieved maximum mechanical properties.

Keywords—Metakaolin, geopolymer, polymer admixtures, mechanical properties, microstructure.

I. INTRODUCTION

NOWADAYS, the alkali-activation is a globally growing technology that involves the chemical reaction between a solid aluminosilicate precursor and an alkaline activator, at room temperature, giving a hardened product. Investigation in the field of alkali-activation had an exponential increase according to the research results by the French author Davidovits [1] who developed and patented binders obtained from the alkali-activation of metakaolin, having named it using the term ''geopolymer'' in 1978.

Geopolymers referred to as alkali-activated aluminosilicate binders and comprise three classes of inorganic polymers that, depending on the ratio of silica to alumina (silica/alumina), are based on the following three different monomeric units: (–Si–O–Al–O–), polysialate, $SiO_2/Al_2O_3=2$; (–Si–O–Al–O–Si–O–), polysialatesiloxo; $SiO_2/Al_2O_3=4$;(–Si–O–Al–O–Si–O–Si–O–), polysialatedisiloxo, $SiO_2/Al_2O_3=6$ [2]. Moreover, there are numerous advantages of these alkali activated aluminosilicates system as lower heat of hydration [3], the development of earlier and higher mechanical properties [4], [5], low heat release [6], better resistance against chemical attack [7], [8], freeze-thaw resistance [9], fire resistance [10], [11], higher reduction in chloride diffusion [12] and stronger aggregate-matrix interface formation [13], [14]. However, the alkali-activated aluminosilicate system presents some problems such as rapid setting periods [15], higher shrinkage values [16], and higher formation of salt efflorescence [5], faster carbonation [17] and tendency to crack during curing [18], [19].

Researchers have been trying to solve some of these problems by using different additives in the AAA system. Therefore, by using various types of polymer admixtures it is possible to overcome these problems.
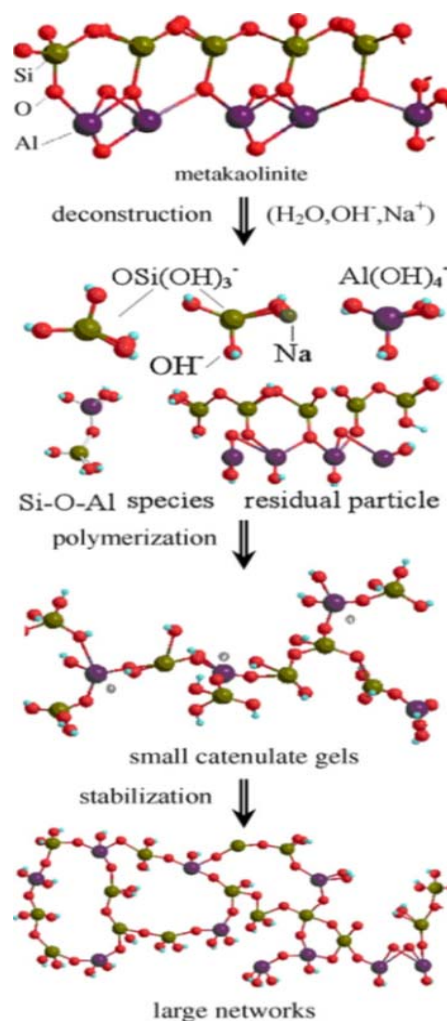

Fig. 1 Sketch of the geopolymerization process, including the deconstruction of MK by the activation of alkaline solution [20]

Geopolymerization is a complicated process responsible for the formation of geopolymer. From the perspective of the changes in response to the reaction heat, the geopolymerization stages of alkali–metakaolin can be split into three stages: I deconstruction, II polymerization and III stabilization as shown in Fig. 1. However, these stages can

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World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:10, No:12, 2016

hardly be separated clearly for they may occur simultaneously [20].

The mechanism by which the geopolymer structure forms is thought to involve three major stages. The first of the three stages involves the depolymerization of the existing poly(siloxo) layer of the kaolinite. This is followed by the formation of $(OH)_3$-Si-O-Al-$(OH)_3$. Finally, there is polycodensation to higher oligomers and polymers that come together to make up the overall structure.

II. EXPERIMENTAL

A. Materials

Metakaolin Mefisto K_{05} (the company CLUZ) was produced through controlled thermal processes and grain size adjustments of clay stones and floating kaolin clays. It is a highly active pozzolan on base of metakaolinite. Its chemical composition is given in Table I. Particle size are $d_{50} = 6.34$ μm and $d_{90} = 11.62$ μm. Sodium water glass with $SiO_2/Na_2O = 1.6$ was used as alkaline activator of metakaolin. Quartz sand with a maximum grain size of 2.5 mm was used as aggregate.

TABLE I
CHEMICAL ANALYSIS OF METAKAOLIN MEFISTO K_{05} [% W/W]

SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	Na_2O	K_2O	LOI
55.01	40.94	0.55	0.55	0.14	0.34	0.09	0.60	1.57

Four types of commercial polymer admixtures were used: VINNAPAS® 5023 L; VINNAPAS® 5111 L; VINNAPAS® 7016 F; VINNAPAS® 7220 E. The first two are polymer powders based on vinyl acetate and ethylene. VINNAPAS® 7016 F is a plasticizing dispersible polymer powder based on vinyl acetate, ethylene and methyl methacrylate. VINNAPAS® 7220 E is a semi flexible dispersible polymer powder based on vinyl esters, ethylene and acrylic acid ester. Also, carboxymethyl cellulose (CMC), hydroxypropyl methylcellulose (HPMC) and polypropylene glycol (PPG) were used. CMC is cellulose derivative and water soluble polymer. The water-solubility is achieved by introducing carboxymethyl groups along the cellulose chain, which makes hydration of the molecule possible. HPMC is a chemically modified cellulose polymer. Geopolymer mortars were produced by mixing 350 g of metakaolin, 350 g of sodium silicate solution, 120 ml of water and 1350 g of aggregates. Each type of VINNAPAS® was added at dosages of 0.5, 1, 1.5, and 2% by mass of metakaolin. CMC and HPMC were added at dosage 0.1, 0.5 a 1% by mass of metakaolin. PPG was added at dosage 1 and 2% by mass of metakaolin. The activator and polymer admixture were dispersed and partially dissolved in water prior to mixing with metakaolin. Then quartz sand was added into the mixture to prepare fresh mortar.

Geopolymer mortar specimens were cast in prismatic moulds: 40 × 40 × 160 mm). The specimens were left in the moulds for 2 hours, then cured in oven at a temperature 40 ºC for 4 hours in order to accelerate the hardening process, and finally removed from the moulds after 24 hours. After demoulding, the specimens were stored in plastic bags for 27 days at RH = 45 ± 5%.

B. Tests

Flexural strength was determined using a standard three-point-bending test, while compressive strength was measured on far edge of both residual pieces obtained from the flexural strength test. Porosity measurements (mercury intrusion porosimetry) were performed only on reference specimen and specimens with the highest and lowest strength. The microstructure of geopolymer mortars was investigated with scanning electron microscope (SEM) (Tescan MIRA3 XMU), applying acceleration voltage of 30 kV. The samples used were dried and sputtered with gold.

III. RESULTS AND DISCUSSION

The results of each test are presented separately and their comparison has been done in a way to study the effect of each type of polymer content.

A. Mechanical Properties

Compressive and flexural strength of the reference metakaolin-based geopolymer mortar (REF) and of the polymer-modified geopolymer mortars by different types of VINNAPAS® are given in Figs. 2 and 3 respectively. The addition of all types of VINNAPAS® resulted in lower compressive strength values compared to REF composition, with the exception of VINNAPAS® 7016 F whose addition contributed to compressive strength increase. The maximum compressive strength was observed for the composition with 0.5% of VINNAPAS® 7016 F, being by 12% higher than that of REF. The composition containing 2% of VINNAPAS® 8031 H showed the minimum compressive strength value, which was by 60% lower than that of REF.

The mortars containing VINNAPAS® 5111 L, 7220 E and 5023 L showed higher flexural strength values than those made with 8031 H; however, only the specimens containing VINNAPAS® 7016 F had flexural strength values higher than the REF specimen. The mortars with 0.5 and 1% addition of VINNAPAS® 7016 F showed 10% higher flexural strength in comparison to the REF one. It should be noted that no efflorescence was observed on the surfaces of the samples with VINNAPAS® 7016 F (Fig. 6), whereas it was present on the other samples, which may explain its high strength.

Compressive and flexural strength of the REF metakaolin-based geopolymer mortar and of the polymer-modified geopolymer mortars by CMC, HPMC and PPG are given in Figs. 4 and 5, respectively. The addition of HPMC and PPG showed the lowest values for compressive and flexural strength. More specifically, the addition of PPG has not any effect on compressive and flexural strength. Nevertheless, intensive efflorescence was formed on the surfaces of the samples with 1 and 2% of PPG. It can explain the lowest mechanical strengths of these samples.

0.1% addition of CMC contributed to compressive strength slightly increase. And the amount of 0.5 and 1% resulted the same values as REF values of flexural strength.

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:10, No:12, 2016

B. Microstructure

Micrographs of the samples obtained from the mortars with VINNAPAS® admixtures showed that these mortars contain plenty of lamellar particulates of approximate mean size of 5 μm (Figs. 8 and 9). It should also be noted that metakaolin particles in REF mortar (Fig. 8) reacted to greater extent than those of the other mortar (Fig. 9). However, the structure of geopolymer mortar with 1.5% 7016F (which had the maximum compressive strength) is more compact, which may explain its high strength.
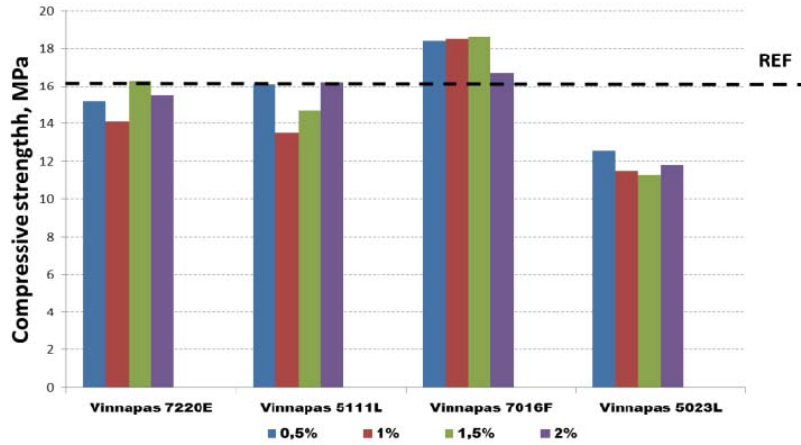
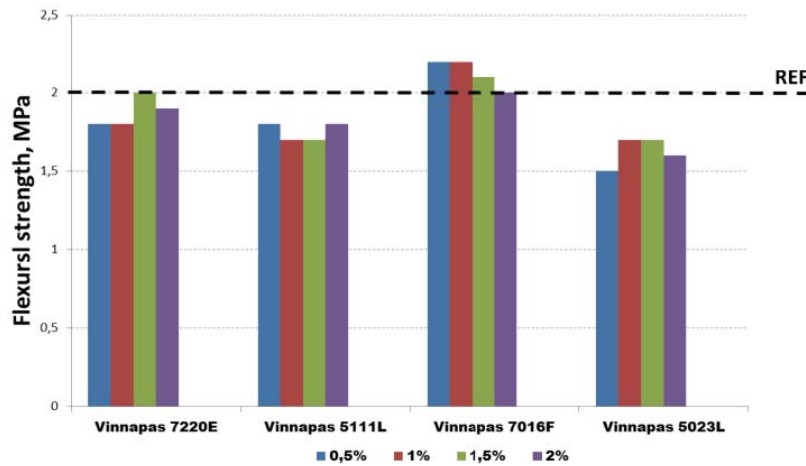Fig. 2 Compressive strength of geopolymer mortars with different contents of different types of VINNAPAS®

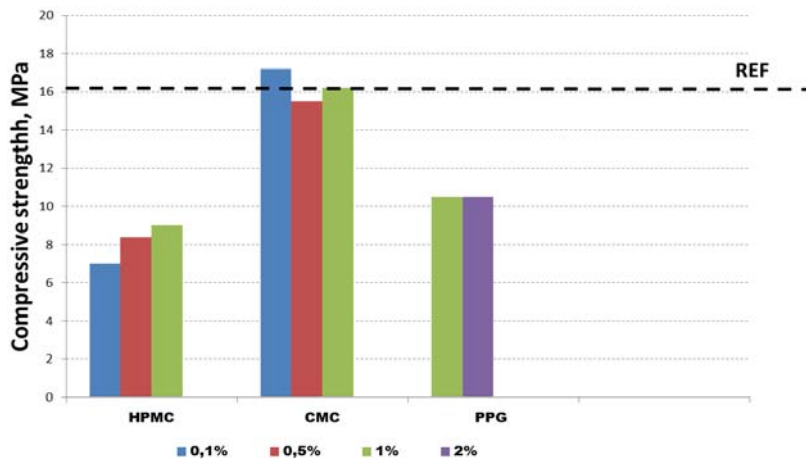Fig. 3 Flexural strength of geopolymer mortars with different contents of different types of VINNAPAS®

Fig. 4 Compressive strength of geopolymer mortars with different contents of HPMC, CMC and PPG

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International Journal of Chemical and Molecular Engineering
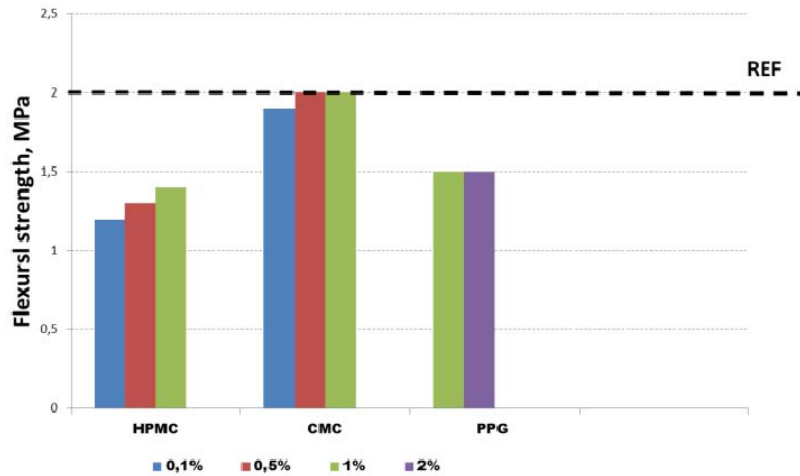Vol:10, No:12, 2016

Fig. 5 Compressive strength of geopolymer mortars with different contents of HPMC, CMC and PPG

Fig. 6 Geopolymer mortar specimens containing 1% VINNAPAS®
7016 F

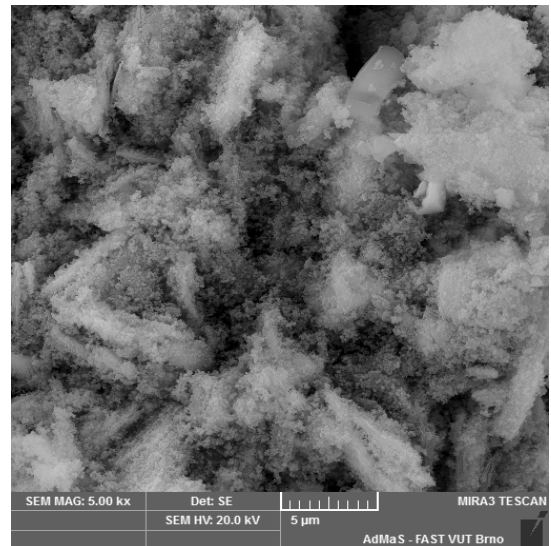Fig. 7 Geopolymer mortar specimens containing 1 and 2% of PPG

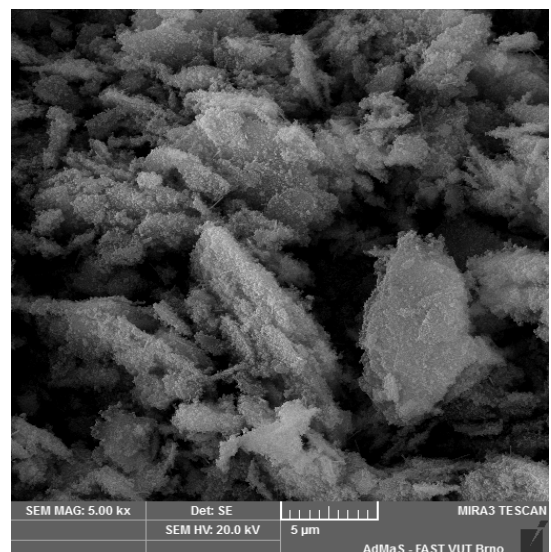Fig. 8 SEM photos (5000× magnification) of REF geopolymer mortar

Fig. 9 SEM photos (5000× magnification) of geopolymer mortars
with 1.5% VINNAPAS® 7016F

World Academy of Science, Engineering and Technology
International Journal of Chemical and Molecular Engineering
Vol:10, No:12, 2016

IV. Conclusion

This paper presents an experimental study on the effect of commercial admixtures on metakaolin-based geopolymers. Several tests were carried out: mechanical (flexural and compressive strength), and microstructural (SEM).

The maximum compressive and flexural strengths were obtained with admixture VINNAPAS® 7016 F for 1.5% and 1% content, respectively. The minimum value of compressive and flexural strength was found for the specimens with HPMC and PPG at dosage 2%. The other types of VINNAPAS® showed slightly decreased mechanical characteristics.

There are very few studies about the effect of polymer admixtures on metakaolin-based geopolymers. Therefore, it is necessary to expand studies in this field of science.

Acknowledgment

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