The Role of Water in the Synthesis of Fly Ash-Based Geopolymer

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Abstract

Water has played an important role in controlling geopolymerization and thus mechanical properties of final geopolymer products. Effects of internal water and atmospheric water on formation of fly ash-based geopolymers and their properties were systematically studied in this research. The initial water amount was varied from 29 to 44 wt%. To investigate the effect of the atmospheric water, curing was designed into 3 successive steps. The samples were first activated at 60 °C in a water saturated-atmosphere for 24 h. Then they were cured at 40 °C for 3 days in two atmospheres; water saturated- or open, prior to curing at 40 °C in an open atmosphere for another 3 days. Microstructure was examined using a scanning electron microscope. Porosity and apparent density were measured following ASTM C 642-06. The results showed that porosity of the samples increased with increasing the initial water content. The initial water content, however, had only minor effect on the apparent density. Curing in the water saturated-atmosphere resulted in a significantly improved compressive strength possibly due to the milder water evaporation. Compressive strength increased with decreasing the initial water contents according to lowered porosity contents. In conclusion, the samples with 29 wt% water content cured in the water saturated - atmosphere showed the highest compressive strength of 34 MPa. This geopolymer paste showed promising potential in construction industry as a cement paste replacement.

Keywords : Fly Ash Based Geopolymer, Compressive Strength, Apparent Density

1. Introduction

Geopolymer is a kind of inorganic polymer made from silica and alumina sources activated by alkaline liquid medium. Geopolymer is considered as a potential alternative construction material to Ordinary Portland Cement (OPC), according to its excellence mechanical properties as well as heat and corrosion resistance. Fly ash has been a major source for geopolymer synthesis due to its high amount of active silica and alumina. Furthermore, fly ash is a by-product from coal combustion so it is abundant and inexpensive. Activation agents mostly used in the geopolymer synthesis were sodium hydroxide and sodium silicate solutions.

In the past decades, attempts to improve mechanical properties, especially compressive strength, of fly ash-based geopolymers have been performed extensively. There are many factors that could affect the compressive strength of fly ash-based geopolymers. The finer fly ash particles giving rise to higher compressive strength was reported (Bohlooli, 2012). The optimum concentration of sodium hydroxide solution was mostly ranged between 10 and 15 M (Chindaprasirt, 2007; Panias, 2007; Ayuso, 2008; Chindaprasirt, 2009; Rattanasak, 2009; Somaratna, 2010; Nazari, 2011; Arioz, 2012; Bohlooli, 2012; Joseph, 2012; Rajamma, 2012; Tho-in, 2012; Ryu, 2013; Vora, 2013 & Görhan, 2014). The best weight ratio of sodium hydroxide to sodium silicate was 1:1 (Chindaprasirt, 2007; Rattanasak, 2009; Rajamma, 2012 & Ryu, 2013). Some works showed the relationship between the compressive strength and the curing temperature. The most appropriate curing temperature reported was in the range of 20 - 90 ^{o}C (Jaarsveld, 2002; Ayuso, 2008; Temuujin, 2009b; Sukmak, 2013; Vora, 2013 & Xie, 2014).

Initial water content was another factor that could significantly affect the mechanical properties of geopolymers. Water is a reaction medium for ions transportation during geopolymerization. After geopolymerization, some water forms hydroxyl, while the excess water is expelled from the three-dimensional network. Increasing initial water content to beyond the needed amount could thus cause a reduction of compressive strength (Temuujin, 2009a; Ferone, 2011 & Vora, 2013). Curing condition was also found to play an important role in controlling the mechanical properties. Aggressive water evaporation could result in cracking and thus a diminish of significant properties of

material. The work by Jaarsveld showed that a higher compressive strength was obtained from the samples cured in open air (Jaarsveld, 2002). It was, however, later reported that the higher compressive strength was achieved with a closed curing setup (Izquierdo, 2010). The best curing regime is thus still controversial.

This work was thus aimed to systematically study effects of the internal water content and curing atmospheric water on formation of fly ash-based geopolymer and their respective properties. To improve the mechanical properties, curing condition was re-designed and addressed in this report.

2. Materials and Methods

2.1 Materials High calcium fly ash (FA: Mae Moh, Lampang, Thailand) was used as silica and alumina sources in this study. Sodium hydroxide (NaOH) solution with a concentration of 10 M and sodium silicate solution was used as the activating agents. The chemical composition of FA, determined using an X-ray fluorescence spectrometer (HORIBA, MESA-500W), is shown in Table 1. The particle size distribution, determined using a Laser diffraction particle distribution analyzer (Malvern, Mastersizer 2000, Hydro 2000MU), is shown in Table 2. The chemical composition of Na_2SiO_3, provided by the supplier (C. THAI CHEMICALS CO., LTD), is shown in Table 3.

Table 1. Chemical composition of fly ash determined using an X-ray fluorescence spectrometer

FA	SiO_2	CaO	Fe_2O_3	Al_2O_3	SO_3	K_2O	TiO_2	Mn_2O_3	BaO
Composition (wt%)	29.765	24.545	19.074	14.248	9.226	2.309	0.526	0.183	0.124

Table 2. Particle size distribution of fly ash determined using a Laser diffraction particle distribution analyzer

Distribution	D10	D50	D90	Average
Size (μm)	2.23	26.56	134.93	54.67

Table 3. Chemical composition of sodium silicate as supplied by the manufacturer

Na_2SiO_3	Na_2O	SiO_2	H_2O
Composition (wt%)	15.50-17.50	34.25-36.25	46.25-50.25

2.2 Samples preparation Sodium silicate and 10 M NaOH solutions were mixed together at a weight ratio of 1:1 for 3 min to form an activating solution. Fly ash, at a fly ash to solution weight ratio of 1:0.6, was added into the activating solution under stirring for 30 min. The water content selected was 29, 34 and 44 wt%, with 29% as the lowest water content possible for paste fabrication. To obtain the greater initial water contents, extra water was added during solid/liquid mixing. The mixture was poured into a cylindrical shaped-plastic mold of 11.6 mm-diameter and 29 mm-height. Curing was performed in three successive steps. The samples were first kept in a sealed container together with 180 mL-water in a separated cup to generate the water saturated-atmosphere. The sample sets were kept in an oven at 60 ℃ for 24 h. In the second step, one set of the samples was further cured in the same container while the other was cured in an open atmosphere, both at 40 ℃ for 3 days. Finally both sets were cured at 40 ℃ in an open atmosphere for another 3 days. The notation of samples i.e. water saturated or open was according to the second step curing.

2.3 Compressive strength test Compressive strength was determined using a universal testing machine (INSTRON 5566) after 7 days. The loading speed was set at 1.0 mm/min and the maximum load of 10.0 kN was used. Average value was obtained from five samples.

2.4 Microstructure, porosity and apparent density Scanning electron microscope, SEM, (LEO 1450 VP) was used for microstructural examination. For porosity and apparent density determination, the samples were dried in an oven at 105 ℃ for 24 h, and then immersed in water at an ambient temperature for 24 h. Vol. of voids and apparent density were measured following ASTM C 642-06. The average value was obtained from five samples.

3. Results and Discussion

3.1 Effect of curing condition on the compressive strength Fig.1 showed that at the same initial water content, the samples cured in water saturated-atmosphere in the second step gave a significantly higher compressive strength than those cured in the open atmosphere. The samples with an initial water content of 29 wt% had the compressive strength of 34 MPa which was around 3 times greater than that obtained from the samples cured in open air. The value increased from 7 to 30 MPa which was about 4 times in the sample sets with an initial water content of 34 wt%. However, the difference was not remarkable for the samples with an initial water content of 44 wt%.

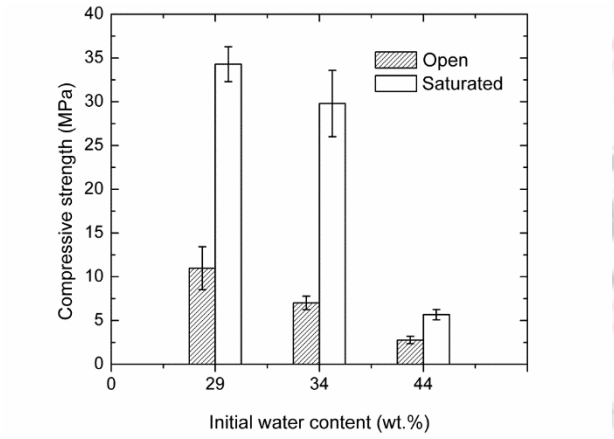

Figure 1. Effects of curing conditions in the second step on compressive strength of geopolymer samples

3.2 Effect of initial water content on compressive strength, apparent density and porosity The relationship among compressive strength, apparent density, porosity and initial water content of fly ash-based geopolymer was clear under open curing condition. The compressive strength decreased significantly, from 11 to 3 MPa, while the porosity was increased from 24.7 to 32.2 vol%, with increasing initial water contents from 29 to 44 wt% (Fig. 2). Therefore, it was clear that under open curing condition, the compressive strength decreased with increasing initial water contents as a result of increasing porosity. It was also shown that the apparent density remained almost constant, regardless of different initial water contents.

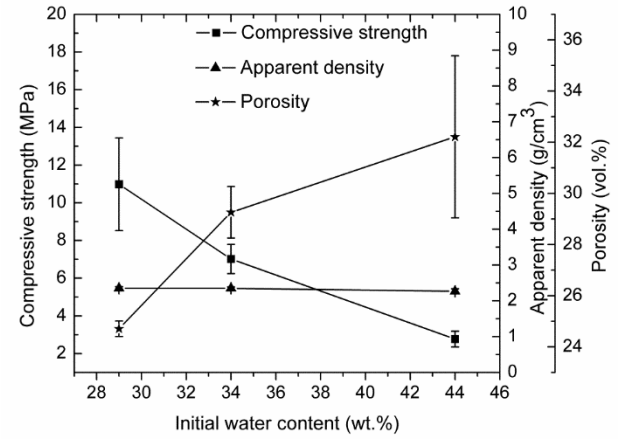

Figure 2. Effects of initial water contents on compressive strength, apparent density and porosity of geopolymer samples cured in open atmosphere in the second step

In an open atmosphere, a similar trend was also obtained, as shown in Fig. 3. The compressive strength dropped sharply from 34 to 6 MPa while the porosity increased from 25.7 to 37.2 vol% with increasing initial water contents from 29 to 44 wt%, while the apparent density was almost unchanged.

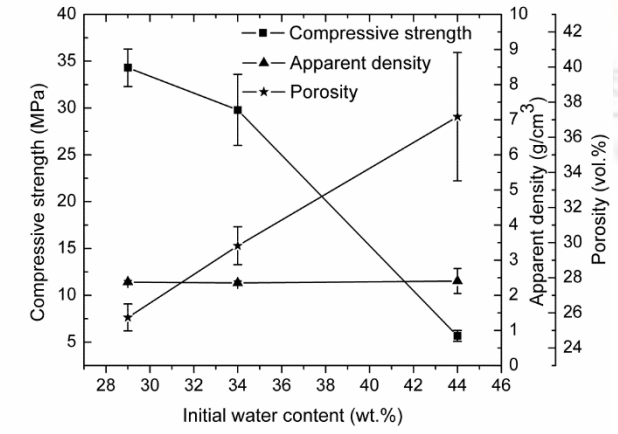

Figure 3. Effects of initial water contents on compressive strength, apparent density and porosity of geopolymer samples cured in water saturated-atmosphere in the second step

3.3 Scanning electron microscope analysis SEM micrographs for the samples with two initial water contents of 29 wt% and 44 wt% were comparatively shown in

Fig. 4 and Fig. 5 respectively. Surface cracks were observed in both geopolymer samples suggesting the great amount of water evaporation during curing. According to Fig. 4(a) and Fig. 5(a), the samples with an initial water content of 29 wt% showed a much smoother surface than the samples with 44 wt% water content suggesting a possible higher degree of reaction progress. Porosity of the samples with 29 wt% water content was obviously less (Fig. 4(b) and Fig. 5(b)) than the ones with 44 wt% water content.

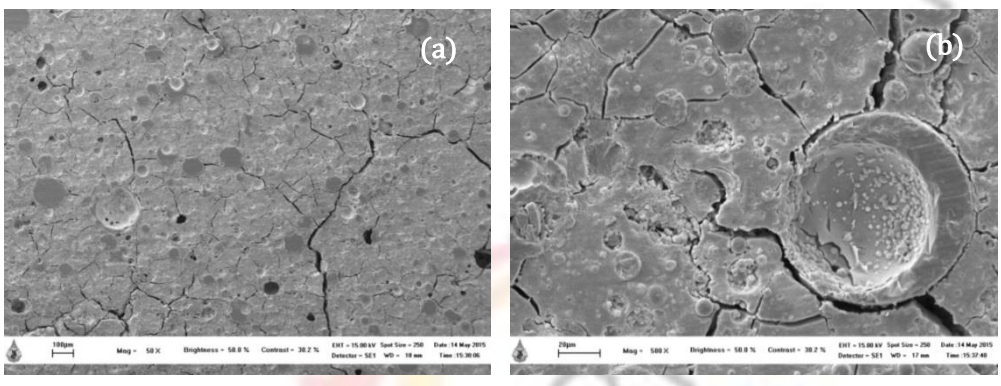

Figure 4. SEM images on polished surface of the geopolymer sample prepared with an initial water content of 29 wt% in water saturated-atmosphere

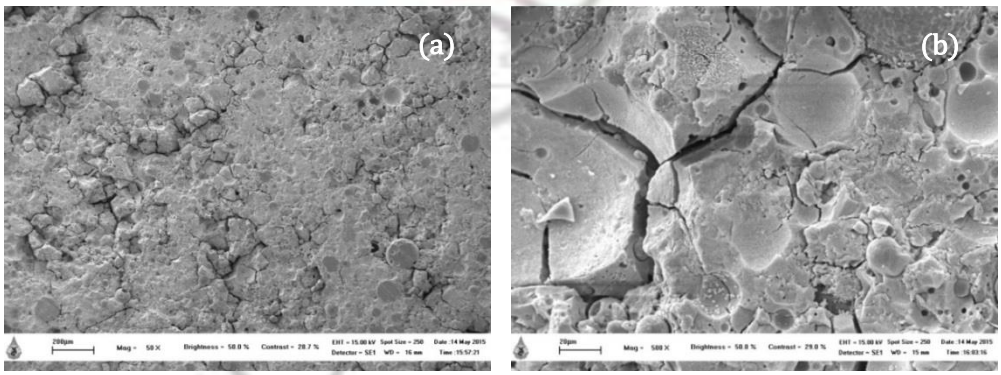

Figure 5. SEM images on polished surface of the geopolymer sample prepared with an initial water content of 44 wt% in water saturated-atmosphere

4. Discussions

Significantly higher compressive strength could be obtained from the geopolymer samples produced in the water saturated atmosphere in the second step. However, at the same water content, the geopolymer samples cured differently showed almost the same

apparent density but slightly different porosity. The samples cured in an open atmosphere showed a slightly greater porosity level. It was likely that porosity played some role, but not dominant, in controlling of the compressive strength. The role of water at the early stage of polymerization could be so significant that improvement of the compressive strength was clearly observed in the samples cured in the water saturated atmosphere. Water played a role as the medium for transportation of the reacting ions so the greater degree of polymerization could be obtained in the samples with sufficient amount of water during that stage. Further examination is required to clarify this statement.

Compressive strength of the geopolymer samples increased significantly, while the porosity decreased remarkably with decreasing initial water content. Evaporation of excess water could possibly occur more mildly, and the reaction progressed more completely in the samples with lower water contents.

5. Conclusion

Our present research found that water has played highly important role to the properties of fly ash based-geopolymer. Curing in the water saturated-atmosphere could greatly improve the compressive strength of geopolymer samples, indicating effectiveness of the proposed curing setup. Compressive strength was increased with decreasing initial water content in the experimental range. This trend was observed in both curing conditions.

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7. References

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