

EFFECT OF Na_2SiO_3 ON COMPRESSIVE STRENGTH OF WASTE GLASS REINFORCED METAKAOLIN-BASED GEOPOLYMER

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Abstract

In comparison with other construction materials such as aluminium, steel, and lumber, Portland cement provides several advantages in terms of ecology and technology. The traditional production process of Portland cement usually generates a large amount of CO_2 during decomposition; moreover, a great deal of fuel is consumed for the high temperature process. Geopolymer was introduced to compensate in cement-based concrete materials because of its low emission of CO_2 and suitable strength. In this study, calcined clay mineral from Lampang province was used as a geopolymer-based material. Prior to utilization, the clay was calcined at 700°C for 2 h to ensure a suitable condition for synthesis of the metakaolin phase. An amount of 30 wt% of waste glass was studied as a possibility for the reinforcement phase in the metakaolin-based geopolymer. In addition, 10 M of NaOH solution and partial NaOH replacement by 30-90 wt% of Na_2SiO_3 were used as the alkaline activator. Samples were cured at 60°C for 7 days. Phase investigation, microstructure, and compressive strength were examined by X-ray diffraction, scanning electron microscope, and universal testing machine, respectively. The results show that the compressive strength decreased from 36.83, 14.00, and 20.37 MPa related to the concentration of Na_2SiO_3 from 30, 60, and 90% by weight, respectively. Fourier-transform infrared spectra corresponding with the compressive strength by transformation of Si-O-Al from substitution in tetrahedral sites of Al in the Si site were also investigated. The microstructure revealed that the samples using 30% waste glass and 30% Na_2SiO_3 were homogeneous due to the dissolution of metakaolin.

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Introduction

Over the past century, concrete-based Portland cement has been the most widely-used material in the construction industry due to its several advantages in terms of ecology and technology in comparison with other materials such as aluminum, steel, and lumber (Gomez-Zamorano *et al.*, 2016). Portland cement is derived from silica (SiO_2), alumina (Al_2O_3), and calcium oxide (CaO). The mix compositions are burned at temperatures between 1400 to 1600°C. In addition, other advantages of Portland cement are that it is inexpensive and widely available because the energy required for concrete production is low when compared with steel. Nevertheless, the traditional process of the production of Portland cement generates a large amount of CO_2 during the decomposition process and consumes a great deal of fuel during the high temperature procedure (Hardjito *et al.*, 2004). Although the cement industry has tended to reduce emissions by around 10% per ton of cementitious product between 2005-2014, current economic growth indicates that the demand for Portland cement will increase CO_2 emissions continuously (CEMBUREAU, 2014). However, the challenge for alternative products could be in the field of partial replacement materials such as low energy cement or alkali- activated materials. The development of alternative materials for Portland-based building construction and other applications has been pursued continuously in the past years (Escalante-García, 2015). During the 1970s, Joseph Davidovits discovered a kind of ecologically friendly material with ceramic-like properties. Geopolymers are inorganic aluminosilicate materials which are rich in alumina and silica and are applied as an alternative binder in the cement industry. Geopolymer formation involves a chemical reaction at ambient temperature by activating with alkali hydroxide (Davidovits, 1994). The geopolymeric material can be applied with

reused waste products with cementitious properties such as fly ash or slag, can be used to immobilize toxic waste, and has the ability to be cast into a desired shape (Leong *et al.*, 2016; Onutai *et al.*, 2016). For these reasons, the development of geopolymer has been promoted by using waste materials that contain high amounts of silica to increase sustainability in the geopolymerization process. This aluminosilicate material can achieve high strength and harden at room temperature from the chemical reaction between the amorphous aluminosilicate and alkali metal silicate solution (i.e. sodium hydroxide or potassium hydroxide). Much published research has reported on the improvement in strength by using different raw materials (Wang *et al.*, 2005; Pouhet and Cyr, 2016). Kaolinite has been widely used for the starting material to synthesize the clay-based geopolymer because of the rich Al_2O_3 and SiO_2 composition in the range of 70-90%, depending on the geology of the location of origin (Heah *et al.*, 2012). The calcination process of kaolin at 600-900°C obtains a highly reactive amorphous phase that can synthesize geopolymer and the study of metakaolins has been dominated by the effect of many parameters such as the Si/Al ratio, molar concentration of the alkaline solution, and curing condition of the compressive strength. In the case of a complex silicate source such as waste glass, zeolite, and fly ash there is an effect on the results from impurities in the raw material.

This study presents the results of the alkali activator mixed from sodium hydroxide/sodium silicate in various concentrations and the characterization of the geopolymer materials based on metakaolin partially replaced with 30 wt% waste glass. The purpose was to analyze the effect of mixing the alkali activator binder on the compressive strength.

Experimental Procedure

The starting material used in the solid component as the source of Al_2O_3 and SiO_2 was local kaolinite clay from Lampang province in the north of Thailand. To achieve a pure phase of metakaolin, the kaolinite was calcined at 700°C , with the temperature determined by differential thermal analysis (DTA) for 2 h (Kakali *et al.*, 2001). The phase analysis of the calcined clay was confirmed by X-ray diffraction (XRD) using a D2 Advance (Bruker Corp., Billerica, MA, USA) with $\text{CuK}\alpha$ radiation between 2-theta of 10 – 60° . Regarding the silica source or reinforcement material, waste glass (WG) was washed with water, ground, and milled with a ball mill for homogenization. The dried waste glass was de-agglomerated in a mortar and passed through a 60 mesh sieve before use. The chemical compositions of the metakaolin (MK) and WG were verified using X-ray fluorescence (XRF), as shown in Table 1. As suggested by one of the researcher's previous work, partial replacement of the MK by 30 wt% WG was fixed as a solid component material. The utilization of 10 M of NaOH as an alkali activator yielded the highest compressive strength with the value of 22.01 Mpa (Chokkha *et al.*, 2017). To improve the

compressive strength, the mixing solutions of the 2 alkali activators were studied in this paper. The concentration of 10 M NaOH was fixed. However, a partial replacement of 10 M NaOH by an Na_2SiO_3 solution was tested with the weight ratio of 100:0, 70:30, 60:40, and 10:90, respectively. In addition, the mixing composition of all specimens was fixed with the alkali solution/solid component or powder (S/P) ratio of 0.67. After that, a homogeneous gel was achieved and cast into a cubic mold of 50 mm. Later, all samples were sealed with plastic under a curing temperature of 60°C for 7 days. Once the curing process was complete, cube samples were tested to evaluate the compressive strength development of the geopolymer samples with an Instron machine series number 5528 (Instron, Norwood, MA, USA). To support the result from the compressive strength, the microstructure of the geopolymer samples was investigated using a JSM-6010LV electron microscope (JEOL Ltd., Tokyo, Japan). In addition, all samples were pressed into pellets with KBr for the Fourier-transform infrared spectra with a Bruker machine (Bruker Corp., Billerica, MA, USA) to confirm a functional group of geopolymer samples. The procedure of the mix design methodology for the geopolymer is illustrated in Figure 1.

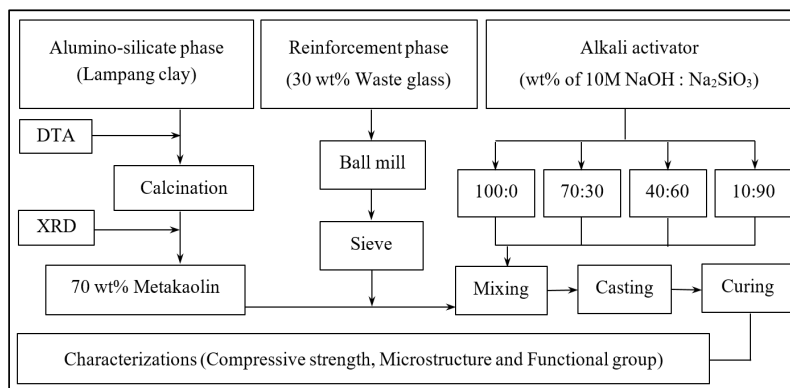


Figure 1. Flow chart of experimental procedure

Results and Discussion

The DTA and derivative curve of the Lampang clay are shown in Figure 2. Endothermic and exothermic peaks were observed in the DTA curve at 610°C and 1015°C, respectively. The endothermic dehydroxylation of chemical water in the kaolinite was observed over a wide range of temperatures. In addition, the onset and endset of the endothermic curve were started around 500°C and stopped around

700°C, respectively. To confirm the results from the DTA, the derivative of the curve was used to examine the loss of the chemical water during the process. The chemical water in the kaolin was decomposed by 1 step at the peak temperature around 610°C. Therefore, the transformation of kaolinite into metakaolin is shown in Equation 1. Regarding another exothermic reaction peak at 1015°C, the result shows the phase transformation of metakaolin into an aluminium-silicon spinel structure

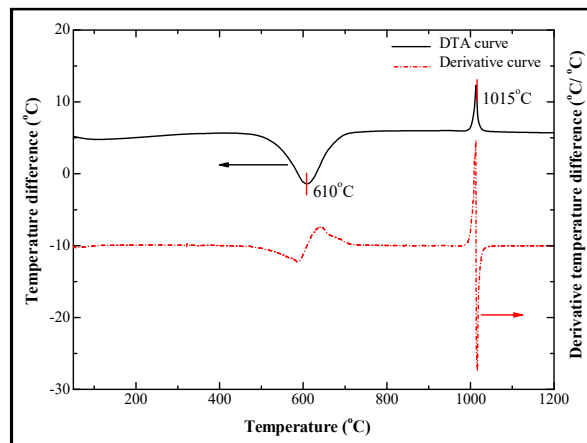


Figure 2. DTA analysis of Lampang clay in the temperature range of 50-1200°C

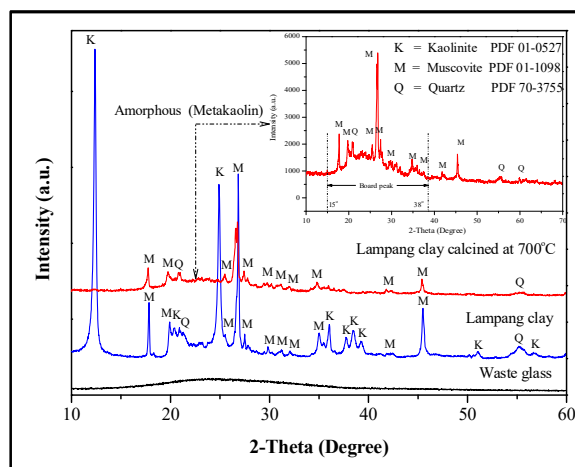
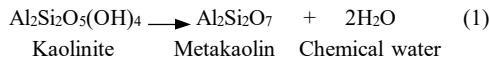


Figure 3. XRD patterns of Lampang clay and waste glass

producing disordered metakaolin and is complete around 650°C. This corresponds with Equation 1 below:



In this study, kaolinite was calcined at 700°C, which is the actual calcination temperature, for 2 h in order to produce a pure phase of metakaolin.

Figure 3 shows the XRD patterns of the starting materials. The WG exhibited the amorphous phase with the hump around 15-38°. The Lampang clay was composed of kaolinite (PDF 01-0527), muscovite (PDF 01-1098), and quartz (PDF 70-3755), respectively. After being calcined at 700°C, the phase transformation of the kaolinite to amorphous metakaolin was indicated by characteristic peaks at 2-theta of 12.5°, and the main peak of the kaolinite disappeared and was changed to the broad peak in the 2-theta range of 15-38°. This result shows that the optimum time and temperature for the calcination of metakaolin was 700°C and 2 h, respectively. However, minor purities of muscovite and quartz were detected in the calcined kaolin.

Figure 4 shows the compressive strength of all the geopolymer samples. The compressive strength of the geopolymer was increased with an increasing concentration of 30 wt% Na₂SiO₃. In this work, the geopolymer specimen with the mixing ratio of 30 wt% Na₂SiO₃ exhibited the highest compressive strength with the value of 36.83 Mpa. However, the lowest value of compressive strength with 14.00 MPa was the 60 wt% Na₂SiO₃. These results demonstrated the effect of the NaOH/Na₂SiO₃ at various ratios on the compressive strength for the metakaolin replacement by waste glass. The results also show that the strength of the samples increased because the Na₂SiO₃ acted as a waterglass liquid for the geopolymerization process and behaves like a binder. In the process of the geopolymerization setting from the mixing, the alkali activator system played the role of the activator and the binder depends on the ratio of the solution. However, the compressive strength of the samples with compositions of 60 wt% and 90 wt% Na₂SiO₃ dropped because the high amounts of Na₂SiO₃ might retain the strength of the geopolymerization network due to the brittle formation of the excessive waterglass liquid. Corresponding with the

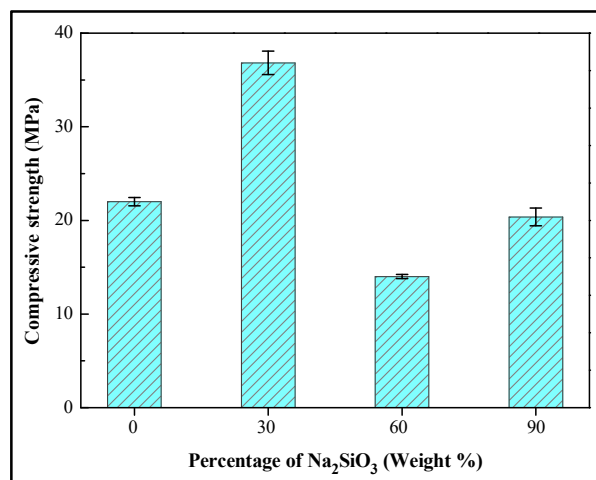


Figure 4. The compressive strength of all geopolymer specimens

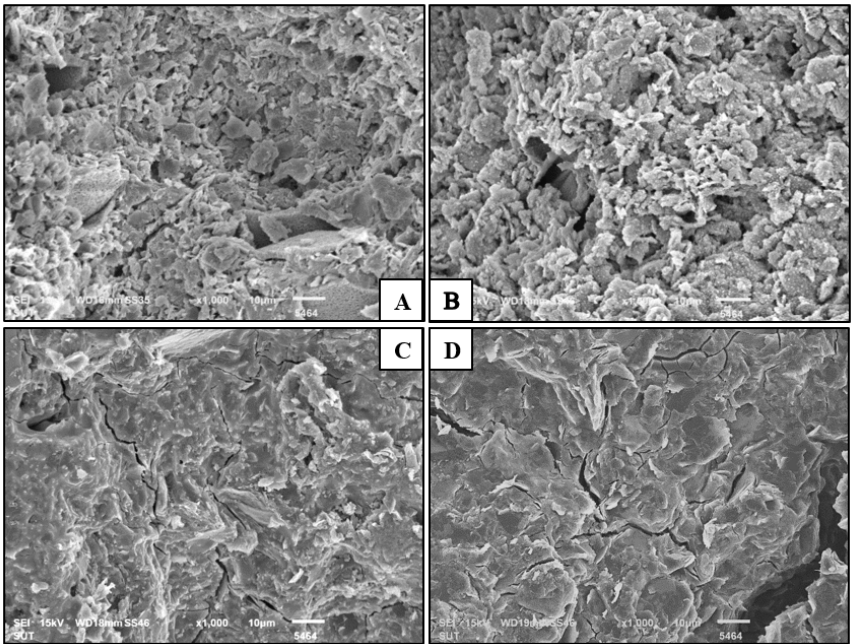


Figure 5. SEM micrograph of the geopolymer samples: (A) 0 wt% of Na_2SiO_3 ; (B) 30 wt% of Na_2SiO_3 ; (C) 60 wt% of Na_2SiO_3 ; and (D) 90 wt% of Na_2SiO_3

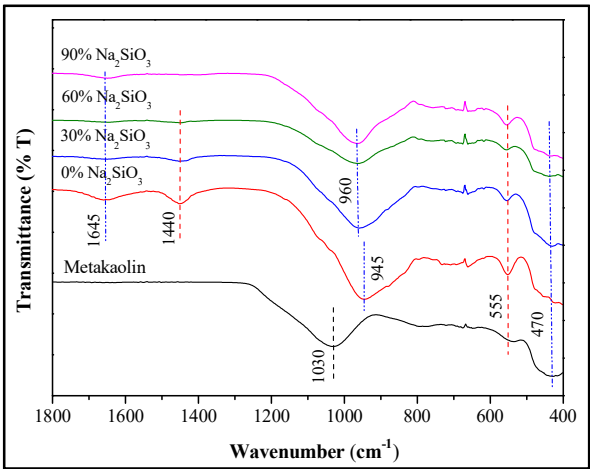


Figure 6. Fourier-transform infrared spectra of the geopolymer samples

microstructure in Figure 5(C) and 5(D) of 60 and 90 wt% Na_2SiO_3 , respectively, both samples were observed with microcracks in the structure and the microcracks' propagation

reduced the compressive strength of the samples. Furthermore, the lower ratio of the NaOH/Na₂SiO₃ concentration affected the strength due to the high viscosity of the Na₂SiO₃ liquid which inhibited the dissociation of the Al and Si into the network of the geopolymer (Heah *et al.*, 2012). Therefore, the metakaolin-based geopolymer requires a suitable NaOH/Na₂SiO₃ ratio because the formation of the geopolymer network depends on dissolution of the metakaolin. An alkali activator with a high NaOH concentration improves the ability to dissolve the metakaolin particles (Wang *et al.*, 2005).

The cross-section microstructure of the samples is shown in Figure 5. Metakaolin reinforcement with 30 wt% waste glass-based geopolymer with the mixing alkali activators of 10 M NaOH and Na₂SiO₃ (100:0 and 70:30 wt% ratios) was demonstrated as a homogeneous and fine microstructure. Therefore, the compressive strength of these geopolymer samples was increased with an increasing concentration of Na₂SiO₃. The 30 wt% Na₂SiO₃ shows the highest compressive strength because of the increasing hard phase of the SiO₂. However, the hard phase of the SiO₂ increased with increasing the concentration of 60 and 90 wt% Na₂SiO₃.

The Fourier-transform infrared spectra of the metakaolin-based geopolymer samples with different NaOH/Na₂SiO₃ ratios are presented in Figure 6. All samples indicated the transformation by geopolymerization process from the transmittance spectra. Accordingly, the main peak of the metakaolin sample around 1030 cm⁻¹ was indicated in the remaining quartz (Akolekar *et al.*, 1997). The transmittance peaks were observed at 555 and 470 cm⁻¹ and refer to the asymmetry of the Si-O-Al and Si-O-Si. For the sample with only NaOH, the transmittance peaks at 1645 and 1440 cm⁻¹ were the O-H bending bond of water and the stretching vibration of the O-C-O bond, respectively. However, the main peak of the NaOH/Na₂SiO₃ ratios (100:0, 70:30, 40:60, and 10:90) was observed at the lower wavenumber (945- 960 cm⁻¹). The result is related to the partial replacement of Si by Al in the geopolymer gel structure and also suggests

that the geopolymer degree strengthened (Duxson *et al.*, 2005).

Conclusions

The effect of Na₂SiO₃ on the compressive strength of metakaolin-based geopolymer was investigated. The actual calcined temperature at 700°C of Lampang clay was used to achieve the pure phase of the metakaolin. The effect of the NaOH/Na₂SiO₃ ratio played an important role to increase the compressive strength due to the presence of the SiO₂ hardness phase in the waterglass liquid. For the Fourier-transform infrared spectra, all the geopolymer samples show the lower wavenumber around 940-960 cm⁻¹ corresponding with the partial replacement of Al in the Si tetrahedral site. The microstructure was revealed to be homogeneous in 30 wt% of Na₂SiO₃ that supports an increase in the compressive strength. However, microcracks appeared in the 60 and 90 wt% of Na₂SiO₃ because the high viscosity of the alkali activator inhibited the dissociation of Al and Si into the network of the geopolymer.

References

- Akolekar, D., Chaffee, A., and Howe, R.F. (1997). The transformation of kaolin to low-silica X zeolite. *Zeolites*, 19(5-6):359-365.
- CEMBUREAU. (2014). The Activity Report. The European Cement Association, Brussels, Belgium, 44p.
- Chokkha, S., Phetnat, P., Chandadi, W., and Srisitthigul, M. (2017). Use of waste glass as a reinforce material in calcined-kaolin based geopolymer. *Key Eng. Mater.*, 751:556-562.
- Davidovits, J. (1994). Properties of geopolymer cements. *Proceedings of the 1st International Conference on Alkaline Cements and Concretes*; October 11-14, 1994; Kiev, Russia, p. 131-149.
- Duxson, P., Lukey, G., Separovic, F., and Van Deventer, J. (2005). Effect of alkali cations on aluminum incorporation in geopolymeric gels. *Ind. Eng. Chem. Res.*, 44(4):832-839.
- Escalante-García, J.I. (2015). Overview of the potential of urban waste glass as a cementitious material in alternative chemically activated binders. *J. Chin. Ceram. Soc.*, 43(10):1441-1448.
- Gomez-Zamorano, L.Y., Vega-Cordero, E., and Struble, L. (2016). Composite geopolymers of metakaolin

- and geothermal nanosilica waste. *Constr. Build. Mater.*, 115:269-276.
- Heah, C.Y., Kamarudin, H., Mustafa Al Bakri, A.M., Bnhussain, M., Luqman, M., Khairul Nizar, I., Ruzaidi, C.M., and Liew, Y.M. (2012). Study on solids- to- liquid and alkaline activator ratios on kaolin-based geopolymers. *Constr. Build. Mater.*, 35:912-922.
- Kakali, G., Perraki, T., Tsivilis, S., and Badogiannis, E. (2001). Thermal treatment of kaolin: the effect of mineralogy on the pozzolanic activity. *Appl. Clay Sci.*, 20(1):73-80.
- Leong, H.Y., Ong, D.E.L., Sanjayan, J.G., and Nazari, A. (2016). The effect of different Na_2O and K_2O ratios of alkali activator on compressive strength of fly ash based-geopolymer. *Constr. Build. Mater.*, 106:500-511.
- Onutai, S., Jiemsirilers, S., Thavorniti, P., and Kobayashi, T. (2016). Fast microwave syntheses of fly ash based porous geopolymers in the presence of high alkali concentration. *Ceram. Int.*, 42(8):9866-9874.
- Pouhet, R. and Cyr, M. (2016). Carbonation in the pore solution of metakaolin-based geopolymer. *Cement Concrete Res.*, 88:227-235.
- Wang, H., Li, H., and Yan, F. (2005). Synthesis and mechanical properties of metakaolinite- based geopolymer. *Colloid. Surface. A*, 268(1-3):1-6.