The Effect of Sodium Hydroxide (NaOH) Solution Concentration on Properties of Geopolymer Paste

Farah Farhana Zainal1,* Nurqistina Sulotoha1, Yusrina Mat Daud1, Mohammad Firdaus Abu Hashim1, Hasri2 and Hartati2

1Center of Excellence Geopolymer and Green Technology (CEGeoGTech), School of Materials Engineering, Universiti Malaysia Perlis (UniMAP), P.O. Box 77, D/A Pejabat Pos Besar, 01000 Kangar, Perlis, Malaysia
2Fakultas Matematika & Ilmu Pengetahuan Alam, Universitas Negeri Makassar, Kampus FMIPA UNM, Parang Tambung, JL. Mallengkeri, Makassar, 90224, Indonesia

Abstract. This research analyzes the effect of sodium hydroxide (NaOH) solution concentration ranging from 8M to 12M as alkaline activator on the properties of geopolymer paste. Alkaline activator is essentially a mixture of sodium hydroxide and sodium silicate solution. In this research, the main component used was raw kaolin. In order to produce kaolin based geopolymer paste, the alkaline activator solution was prepared with 0.24:1.00 ratio of Na2SiO3/NaOH and this alkaline activator solution was prepared for 24 hours prior before used in another process. The solid-to-liquid ratio which is kaolin-to alkaline activator solution ratio was 0.80:1.00. The mixture of kaolin based geopolymer paste were placed in cube moulds with a size of 50x50x50 mm, and left for 24 hours until it hardened. Then, the samples were cured at 80°C in the oven for 24 hours. The samples of kaolin based geopolymer paste were tested based on compressive strength, morphology analysis, water absorption and porosity after 28 days. In this project, 8M concentration of NaOH solution was the best concentration in order to study the synthesis of kaolin based geopolymer paste as the sample had the highest amount of compressive strength with 0.992 MPa and had the lowest water absorption and porosity with 1.246% and 24.08% respectively. Scanning Electron Microscope (SEM) was used to observe the morphological structure of the kaolin based geopolymer paste. The sample with 8M concentration of NaOH solution shows that least amount of unreacted particles. The structure of kaolin based geopolymer paste was more dense at 8M concentration as the size of pore decreases. The kaolin based geopolymer paste is suitable for use in the construction building industry as a finishing product such as coating.

*Corresponding author: farahfarhana@unimap.edu.my
1 Introduction

Geopolymer is an inorganic material and inorganic polymers formed by activating silica-aluminum rich minerals such as kaolinitic clay, metakaolin and fly ash mixed with the alkaline hydroxide solution and alkaline silicate solution at ambient conditions or higher temperature level [1-4]. Geopolymer is a new generation of the inorganic substances that differ in microstructure and chemical composition compared to Portland cement [5]. In this study, kaolin was used as the sources of alumino-silicate. Many geopolymer synthesis studies have been conducted using fly ash as the source materials, so in previous research there is lack of information on the use of kaolin in geopolymer synthesis. Kaolin was chosen to see how much kaolin in geopolymer synthesis can stand alone. Concentration of sodium hydroxide (NaOH) solution and curing regime are the important parameters to consider when designing for a particular application of kaolin-based geopolymer [6]. The major problem with this type of study is that not many researchers have conducted studies on the effects of NaOH solution concentration on metakaolin or calcined kaolin, so little information is available regarding the effect of NaOH solution concentration on kaolin-based geopolymer paste [7-9].

Other than that, it is important to investigate the various variables affecting the strength and durability of the kaolin geopolymer paste. It is necessary to identify the variables that have the significant effect on the sample so that the proper solution can be made to avoid the limitations. The two variables that can affect the strength and durability is the concentration of NaOH and curing temperature [10-12]. The solution of NaOH mixed as an alkaline activator with Na₂SiO₃ solution will affect the properties of the kaolin-based geopolymer. The porosity and water absorption of kaolin-based geopolymer paste is very important to measure, as the highest percentage of porosity and water absorption will affect the strength of the sample. Several studies have produced estimates of porosity on geopolymer [5, 6], but there are still insufficient data on kaolin-based geopolymer paste for porosity and water. Different NaOH concentrations was used in this project to investigate the effect of NaOH solution concentration in term of physical and mechanical properties of kaolin-based geopolymer paste.

2 Experimental Procedures

2.1 Materials and Sample Preparation

Kaolin which is an industrial waste acts as a source of aluminosilicate in this study. The kaolin was obtained from a local station. Table 1 shows the elemental composition of raw kaolin examined using Xray Fluorescence (XRF) analysis. The alkaline activator solution was prepared by mixing NaOH solution with Na₂SiO₃ solution.

| Table 1. Chemical composition of kaolin. | wt (%) | Table 1. Chemical composition of kaolin. | wt (%) |
|----------------------------------------|--------|----------------------------------------|--------|
| SiO₂                                   | 36.5   | MnO                                    | 0.042  |
| Al₂O₃                                  | 37.3   | ZnO                                    | 0.035  |
| K₂O                                    | 2.50   | CaO                                    | 0.031  |
| Fe₂O₃                                  | 2.06   | V₂O₅                                   | 0.028  |
| TiO₂                                   | 0.764  | PbO                                    | 0.019  |
| CaO                                    | 0.284  | Ga₂O₃                                  | 0.015  |
| R₂O₃                                   | 0.185  | Cr₂O₇                                  | 0.012  |
| Rb₂O₃                                  | 0.0762 | ZnO                                    | 0.011  |
| BaO                                    | 0.065  | Y₂O₃                                   | 0.0099 |
| Eu₂O₃                                  | 0.063  | Ra₂O₇                                  | 0.007  |
In this study, NaOH solution was prepared by dissolving specified amounts of sodium hydroxide (NaOH) pellets with distilled water to obtain NaOH concentration in the range of 8M to 12M and cooled down to room temperature for 24 hours. Then, the NaOH solution was mixed with sodium silicate (Na$_2$SiO$_3$) solution to form alkaline activator with a Na$_2$SiO$_3$/NaOH ratio of 0.24:1.00 [13]. For the preparation of kaolin based geopolymer paste, the kaolin powder was mixed with alkaline activator until homogenous using solid-to-liquid ratio of 0.80:1.00. Heah et al., stated that the ratio of Na$_2$SiO$_3$/NaOH and S/L ratio with 0.24:1.00 and 0.80:1.00 gives the optimum compressive strength after 28 days [1, 14]. After the homogenous paste was obtained, the mixture of kaolin based geopolymer paste was placed into 50x50x50 mm cube moulds and cured in the oven at 80°C for 24 hours [15]. All the samples were sealed using a thin plastic layer after the curing process in order to prevent the excess of Na$^+$. 

2.2 Sample Characterization

2.2.1 Compressive Strength Test

Compressive strength tests of all specimens were evaluated according to the ASTM C109/C109M-08 by using the Shimadzu Universal Testing machine. A minimum of 3 samples for each concentration were examined after 28 days in order to evaluate the early strength gain for each concentration.

2.2.2 Morphology Analysis

Morphology analysis was performed by using Scanning Electron Microscope (SEM) JSM-6460 LA JEOL Japan in order to study the morphology of the raw kaolin and kaolin based geopolymer paste at different concentration of NaOH solution. The samples were cut into small pieces and coated using Auto Fine Coater before the SEM analysis.

2.2.3 Water Absorption and Porosity Test

Water absorption test was conducted by immersing a minimum of 3 samples for 24 hours and calculating the average data by using equation (1):

$$\text{Percentage of water absorption} = \frac{\text{Wet weight} - \text{Dry weight}}{\text{Dry weight}} \times 100\%$$

In order to identify the value of porosity of the sample, pycnometer was used to analyze the data. The readings were taken by calculating the average value of the total pore within 3 samples and the data is usually presented in percentage.

3 Results and Discussion

3.1 Compressive Strength

The mechanical properties of kaolin based geopolymer paste were identified by compressive strength testing. The compressive strength testing was conducted after 28 days ensure the optimization of geopolymerization process. Fig. 1 shows the compressive strength of different concentration NaOH solution (8M, 10M and 12M) that was cured at
80℃ after 28 days. It can be seen that the compressive strength slightly decreased from 8M to 12M. Sample 8M shows the highest strength with 0.992 MPa, followed by Sample 10M with 0.946 MPa and Sample 12M with 0.754 MPa.

Sample 8M shows the highest value compared to the other samples due to the water content. At lower NaOH solution concentration, the sample mix has excess mixing water. This water content gives benefits to the geopolymerization process where it helps in the transportation of dissolved ions [6]. Other than that, due to the early age strength at 28 days, sample 8M achieved optimum strength. This is because at 28 days, the reaction is mostly fully reacted as kaolin based geopolymer paste.

![Compressive strength with various concentration of NaOH solution.](image)

### 3.2 Morphology of Geopolymers

Analysis of morphology was observed after 28 days and curing at 80 ℃ for 24 hours. Fig. 2 shows the changes in morphologies of kaolin based geopolymer paste for various NaOH solution concentrations. From Fig. 2 (a), (b), (c) and (d), it can be seen that the kaolin based geopolymer paste particles changed in shapes after 28 days when it changes from plate-like structure to spherical sponge-like structure. Raw kaolin was activated by the alkaline activator with various NaOH solution concentrations and the geopolymerization reaction occurred after mixing the raw kaolin with alkaline activator solution [16, 17].
Fig. 2 displays the morphology of kaolin based geopolymer paste with different concentrations of NaOH solution at constant Na$_2$SiO$_3$/NaOH ratio and S/L ratio with 0.24:1.00 and 0.8:1.00 respectively. Other than that, Fig. 2 also shows that sample 8M contained less amount of unreacted particles compared to sample 10M and sample 12M. Sample 12M shows the largest amount of unreacted particles of raw kaolin. Sample 8M contributes the highest strength with 0.992 MPa and this can be related to the morphology of the sample where sample 8M is more homogeneous compared to other samples as shown in Fig. 2. The morphology of sample 8M has more geopolymeric gel than sample 10M and sample 12M due to the water content that contributes to optimum strength of the sample. This morphology test revealed that sample 12M with the highest concentration of NaOH solution has a larger amount of unreacted kaolin based geopolymer paste as shown in Fig. 2 (d). It can also be observed that sample 12M has less geopolymeric gel that is likely to be caused by the excess Na$.^+$ Sample 10M and sample 12M have the lowest strength due to the incomplete chemical reaction during geopolymerization process between raw kaolin and alkaline activator solution presented by the plate-like structure of kaolin. However, all the samples displayed partially reacted particles and showed large amounts of unreacted particles at 28 days. This is due to the characteristics of the kaolin itself. As the plate-like and layered structure provides low reactivity of the kaolin, the dissolution of kaolin and alkaline activator solution was extremely slow [18]. Kaolin based geopolymer paste does not undergo complete geopolymerization process and is not fully incorporated into the structure.

3.3 Water Absorption and Porosity Test

Fig. 3 presents the relationship between water absorption and porosity for different concentration of NaOH solution in terms of molarity. As illustrated in Fig. 3, the trend of the graph shows that the amount of water absorption and porosity are slightly increased
with increasing NaOH solution concentration. The samples with 8M concentration of NaOH solution shows the lowest amount of water absorption and porosity with 1.246% and 0.422% respectively compared to other samples. As expected, the porosity affected the water absorption of kaolin based geopolymer paste. More water will enter the sample that has higher amount of porosity and will cause damage to the sample such as crack. As the amount of porosity increases, the size of the pore also increases, causing the water absorbed to also increase.

Fig. 3. Water absorption and porosity with various concentration of NaOH solution at 28 days.

Conclusions

Detailed work has been done to achieve the objectives of this study. Referring to the experimental investigation, this paper has highlighted the effect of Sodium Hydroxide (NaOH) solution concentration on the properties of geopolymer paste. Results from this investigation can be concluded as following:

1) The concentration of NaOH solution has significant effect on the compressive strength of kaolin based geopolymer paste.
2) Kaolin based geopolymer paste showed the optimum strength at 0.992 MPa for the samples prepared with 8M concentration of NaOH solution cured at 80°C for 24 hours.
3) The relationship of compressive strength with water absorption and porosity of kaolin based geopolymer paste showed the similar trends.
4) Samples with lower water absorption and porosity yielded optimum strength after 28 days.
5) Sample with 8 M concentration of NaOH solution showed the least amount of porosity and displayed less portion of unreacted particles of kaolin based geopolymer paste. This can be proved by morphology study using Scanning Electron Microscope (SEM).

The authors would like to thank the staffs of Center of Excellence Geopolymer & Green Technology (CEGeoGTech), School of Materials Engineering, Universiti Malaysia Perlis (UniMAP) for their involvement in the research. This work was supported and funded by the Universitas Negeri Makassar Kampus FMIPA UNM, Makassar, Indonesia. The authors also would like to acknowledge the support from the Fundamental Research Grant Scheme (FRGS) under a grant number of FRGS/1/2018/TK06/UNIMAP/02/2 from the Ministry of Education Malaysia.
References

1. A. Nmiri, N. Hamdi, M. Duc, and E. Srasra, “Synthesis and characterization of kaolinite-based geopolymer: Alkaline activation effect on calcined kaolinitic clay at different temperatures,” vol. 8, no. 2, pp. 276–290, 2017.

2. H.D. Rozman, A.R. Rozyanty, L. Musa, G.S. Tay. J Wood Chem Technol 30(2), 152-163 (2010)

3. A.R. Rozyanty, H.D. Rozman, G.S. Tay. Adv. Mater. Res. 264-265, 712-718 (2011)

4. H.D. Rozman, A.R. Rozyanty, G.S Tay, R.N. Kumar. J. Appl. Polym. Sci. 115(5), 2677-2682 (2010)

5. M. T. S. Akelyen, M. EL Achouri, J. Mater. Environ. Sci., 8 (5), 1783–1796 (2017)

6. C. Y. Heah et al., Int. J. Miner. Metall. Mater. 20(3), 313–322 (2013)

7. Ahmad Faris, M., Mustafa Al Bakri Abdullah, M., Nizar Ismail, K., Aida Mohd Mortar, N., Firdaus Abu Hashim, M., Hadi, A. IOP Conf. Ser. Mater. Sci. Eng. 551 (1), 012080 (2019)

8. Hashim, M.F.A., Abdullah, M.M.A.B., Sandu, A.V., Puskas, A., Daud, Y.M., Zainal, F.F., Faris, M.A., Hasri, Hartati, IOP Conf. Ser. Mater. Sci. Eng. 572 (1), 012037 (2019)

9. Abu Hashim, M.F., Abdullah, M.M.A.B., Ghazali, C.M.R., Hussin, K., Binhuussain, M., Omar, M.F. Effect of Glass Reinforced Epoxy (GRE) pipe filled with Geopolymer Materials for Piping Application: Compression Properties (2016) MATEC Web of Conferences, 78, art. no. 01066.

10. Daud, Y.M., Kamarudin, H., Ruzaidi, C.M., Osman, A.F., Al-Bakri, M. (2015) Mater. Sci. Forum 803, 58-62 (2015)

11. Daud, Y.M., Hussin, K., Osman, A.F., Ghazali, C.M.R., Al Bakri Abdullah, M.M., Sandu, A.V. Mater. Plast. 54 (3), 543-545 (2015)

12. Mat Daud, Y., Hussin, K., Ruzaidi, C.M., Osman, A.F., Al-Bakri, M., Binhuussain, M. Mater.Sci. Forum 819,290-294 (2015)

13. S. Ramasamy et al., Rev. Adv. Mater. Sci. 42(1), 83–91(2015).

14. Farah Farhana Zainal, Sahrul Abd Hakim, Yusrina Mat Daud, Mohammad Firdaus Abu Hashim, Mohd Mustafa Al Bakri Abdullah, Hasri and Hartati IOP Conf. Ser. Mater. Sci. Eng. 701 012002 IOP Publishing (2019)

15. S. Pangdaeng, “ Int. J. GEOMATE, 14(46) (2018)

16. H. Kamarudin, a M. M. Al Bakri, M. Binhuussain, C. M. Ruzaidi, M. Luqman, and C. Y. Heah., 10,18–24 (2011)

17. Azhar, N.S.D.M., Zainal, F.F., Abdullah, M.M.A.B. AIP Conf. Proc. 2129, art. no. 020046 (2019)

18. C. Y. Heah et al., Constr. Build. Mater., 35, pp. 912–922 (2012)