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EFFECT OF NaOH SOLUTION ON MECHANICAL PROPERTIES AND MORPHOLOGY OF FLY ASH BASED COLD-PRESSED GEOPOLYMER

Cold-pressing method is beneficial in reducing the porosity of geopolymer matrix, but the low liquid volume will result in unreacted aluminosilicate materials. This problem can be resolved by increasing sodium hydroxide (NaOH) concentration. This paper thus investigates the effect of NaOH concentration on the properties of cold-pressed geopolymer. The fly ash was activated by sodium silicate (Na_2SiO_3) and 10, 12, 14 and 16 M of NaOH solution. The dry mix was compacted with a uniaxial hydraulic press and cured for 7 and 28 days. The specimens were measured by porosity and compressive strength measurements. Scanning electron microscopy (SEM) analysis was performed to determine the microstructure of specimens. The geopolymer was optimized at 14 M NaOH with the highest 28-day compressive strength (109.6 MPa) and lowest porosity (8.1%). The SEM micrographs proved that the geopolymer have a dense and compact microstructure.

Keywords: Cold pressing; NaOH concentration; Porosity; Mechanical properties; Morphology

1. Introduction

The term "geopolymer" was coined by Joseph Davidovits in 1978. Geopolymer is an inorganic polymer obtained due to the alkali activation of aluminosilicate raw materials, such as fly ash, metakaolin, rice husk and bottom ash. The chemical reaction that takes place between alkali activator and aluminosilicate materials is an integrated process called geopolymerization [1,2]. The fabrication of geopolymer allow for the utilisation of industrial waste and secondary product, effectively reduce the mine tailings and industrial by-product piles.

Conventionally, geopolymers are fabricated via casting method wherein a significant amount of alkali activator solution is inevitable. The utilization of highly alkali activator solution give rise to the free water release from the material, which responsible for the progressive shrinkage of the matrix, resulting to cracks initiation. The existence of cracks could impair the strength development and durability of resultant geopolymer. Furthermore, the evaporated of free water leaves micro-pores in the matrix and acts as stress concentration point which susceptible to structure failure during loading. It is well known that the mechanical properties of geopolymer are governed by the porosity of geopolymer matrix, in which geopolymer with a less porous structure has a better mechanical performance.

Previous studies [3-5] have proved that the pressing method successfully lessen the porosity of geopolymer matrix and enhance the strength achievement. Compared to the normal casting method, pressing method requires a lower amount of liquid volume. Forgoing the use of highly liquid volume eliminates the impracticalities of handling, transporting and storing highly corrosive alkaline solutions. Nonetheless, the alkaline solution deficiency could potentially hinder the dissolution of source materials leading plentiful remnant solid particles. These circumstances might interfere with the mechanical properties of pressed geopolymer.

One of the prominent factors determining the dissolution of aluminosilicates materials is its mix proportion. Factors that are of particular importance are solid-to-liquid ratio, NaOH concentration and Na_2SiO_3/NaOH ratio. Previous literatures have done on cast geopolymer proved that increasing NaOH concentration contributes to the dissolution kinetics of the aluminosilicate materials [6,7]. Cold-pressed lightweight geopolymer with strengths of 14.1 MPa was fabricated using fly ash and ordinary Portland cement (OPC) activated by 10 M NaOH solution [8]. However, the incorporation of OPC is environmentally unsustainable due

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to the necessity of limestone calcination at 1400°C. Using hot-pressing method, higher compressive strength (100d = 133 MPa) was acquired by Ranjbar et al. [5] wherein the fly ash based geopolymer was fabricated with 16 M NaOH and Na_2SiO_3. In spite of that, heat source is required, which consumed a high amount of energy and proved more challenging to handle in mass production processes [9].

Yet, very limited efforts have been made to investigate the impact of NaOH concentration on the performances of cold-pressed geopolymer. While the use of limited alkali activator in cold-pressing method could possibly restrict the reactivity of the source materials, the problem could be revamped by adjusting the NaOH concentration. Thus, the current study aims to evaluate the performances of cold-pressed geopolymer with different NaOH concentration. This study contributes a basis understanding on the influence of NaOH concentration on the percentage of porosity, compressive strength, and microstructure of cold-pressed geopolymer in 7 and 28 days. The findings of this research pave way the development of high strength cementitious materials with low alkali activator for tiles, bricks and precast concrete applications.

2. Materials and methods

2.1. Material

Class F fly ash utilized as source material in the preparation of geopolymer was collected from Manjung Coal-fired Power Plant, Perak, Malaysia. Using X-ray fluorescence (XRF) analysis, the chemical composition of fly ash was investigated and enlisted in TABLE 1. The fly ash used mainly composed of SiO_2, CaO, Al_2O_3 and Fe_2O_3. Based on Fig. 1, the fly ash particles have a smooth and spherical morphology.

TABLE 1

Chemical composition of fly ash

Compound	SiO_2	CaO	Al_2O_3	Fe_2O_3	SO_3	K_2O	Others
Mass (%)	36.7	19.1	18.7	17.2	3.0	1.8	3.5

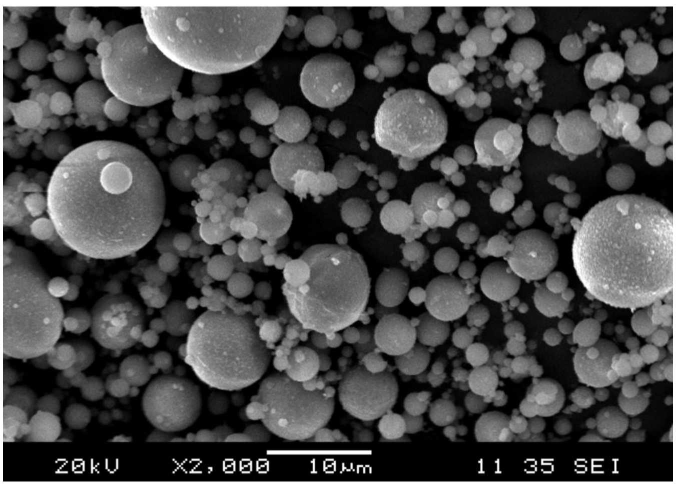


Fig. 1. SEM micrograph of fly ash

NaOH and Na_2SiO_3 solution were used as alkali activator in the fabrication of cold-pressed geoopolymer. The NaOH pellets have a purity of 99.0%, while the Na_2SiO_3 solution has a chemical composition of 30.1% of SiO_2, 9.4% of Na_2O and 60.5% of H_2O with SiO_2/ Na_2O ratio of 3.20.

2.2. Fabrication of geopolymer

In this study, different NaOH concentrations (10, 12, 14 and 16 M) were used to activate the aluminosilicate materials. In order to prepare the NaOH solution, the NaOH pellets were dissolved by distilled water and stored at room temperature for 24 h. The mixing ratio between Na_2SiO_3 and NaOH solution was 1.0, while the alkali activator and fly ash are mixed with fly ash-to-alkali activator ratio of 5.5. The homogeneous mix was poured into a cylindrical stainless steel mould (height = 50 mm and diameter = 25 mm), and then pressed at 5 tons for 2 min. After that, it was removed from the mould and kept at room temperature for 7 and 28 days.

2.3. Testing and analysis

Porosity measurement of cold-pressed geopolymers were carried out as per ASTM C642. The percentage of porosity of geopolymer at 7 and 28 days was calculated using Eq. (1).

$$\text{Percentage of porosity} = \frac{(m_s - m_d)}{(m_s - m_b)} \times 100\% \qquad (1)$$

Where m_s is the saturated weight, m_d is the dry weight and m_b is buoyant weight of specimen.

The 7-day and 28-day compressive strength of geopolymer specimens were determined according to ASTM C39 using Instron Machine Series 5569 Mechanical Tester with a loading rate of 1 mm/min. The average compressive strength of cold-pressed geopolymer was calculated from the three cylindrical samples.

The microstructure of 28-day cold-pressed geopolymer was evaluated by Scanning Electron Microscopy (SEM) model JSM-6460LA. The electron beam energy and working distance were set at 25 kV and 10 mm.

3. Results and discussions

3.1. Porosity analysis

The percentage of porosity of cold-pressed geopolymer with different NaOH concentration after 7 and 28 days are illustrated in Fig. 2. The porosity of geopolymer ranged from 8.1 to 11.1%. Higher porosity (13-18%) was acquired by cast geopolymer [10] ascribed to the higher liquid dosage that prompted to air bubbles to be trapped in the geopolymer slurry during moulding.

The porosity of cold-pressed geopolymer has lowered from 7 to 28 days of curing, irrespective of the concentration of alkali activator. The reduction was due to the continuous geopolymerization reaction over the time which inhibiting the unreacted fly ash and permitting more reaction products. A similar phenomenon was also observed in cast geopolymer in which the prolong curing enhanced the bonding and compactness of geopolymer matrix [11]. For instance, the increased of curing reduced the porosity of fly ash-metakaolin blend cast geopolymer (NaOH concentration = 12 M) by 4% [12]. This justified the time's pivotal role in the geopolymer development that cured at room temperature.

In general, the increased in NaOH concentration from 10 to 16 M resulted in lower percentage of porosity of cold-pressed geopolymer. At sufficiently high NaOH concentration, the dissolution process (Fig. 3) of aluminosilicate materials improved. This consequently increases the degree of polycondensation and compactness of geopolymer. In the case of low concentration (10 M), the amount of OH^- ions was minimized, leading to a lower efficiency of dissolution of Si^{4+} and Al^{3+} ions from fly ash.

It should however be pointed out that, the percentage of porosity increased when the NaOH concentration went higher than the optimum (14 M). The relatively high viscosity of NaOH solution at higher concentration results in less workable and inhomogeneous mixture. This circumstance give rise to the percentage of porosity of resultant geopolymer.

3.2. Compressive strength analysis

The 7-day and 28-day compressive strength of cold-pressed geopolymer fabricated with varied NaOH concentration are presented in Fig. 4. The 7-day strength of geopolymer increases with NaOH molarity, maximizing the compressive strength of 91.8 MPa with 16 M of NaOH. Increasing NaOH concentration improved the alkalinity of the mixture, favouring a higher degree of reaction. The amount of geopolymer matrix synthesized has a decisive impact on the mechanical performance of geopolymer. The outcome was in good agreement to the reduced porosity as NaOH concentration increased (Fig. 2).

Contrarily, the low NaOH concentration restrained the gepopolymerization reaction attributed to the relatively low pH. The low alkalinity inhibited the OH^- ions participated in the dissolution process of Si^{4+} and Al^{3+} ions, which restricted the compressive achievement of geopolymer. The result comply with the hot-pressed geopolymer where the compressive strength was slightly enhanced (about 5%) when the NaOH concentration increased from 8 to 16 M [5]. This indicated that the compressive strength of pressed geopolymer is influenced by the dissolution of aluminosilicate materials which is relatable to the NaOH concentration.

Although the inclusion of 16 M NaOH solution can maximize the strength of geopolymer at 7 days, the strength devel-

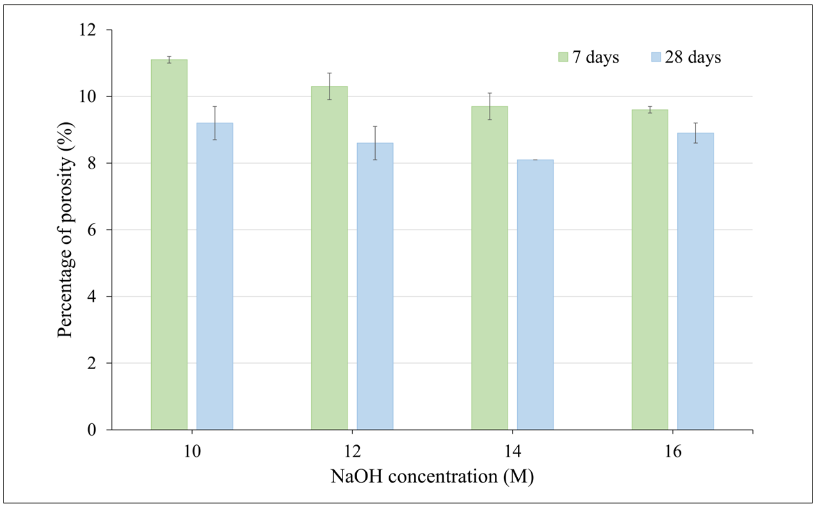


Fig. 2. Percentage of porosity of cold-pressed geopolymer with different NaOH concentration after 7 and 28 days

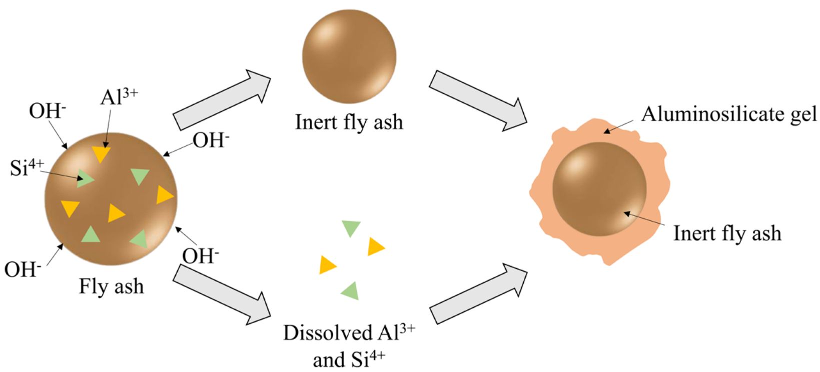


Fig. 3. Dissolution process of fly ash

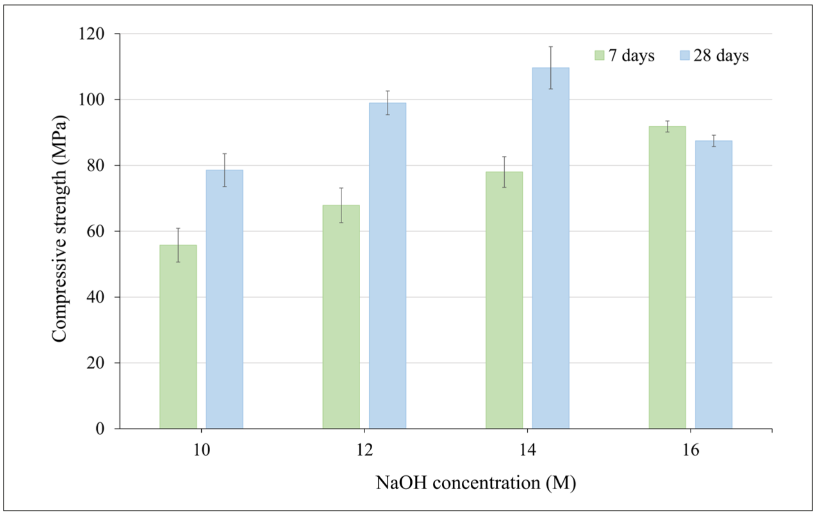
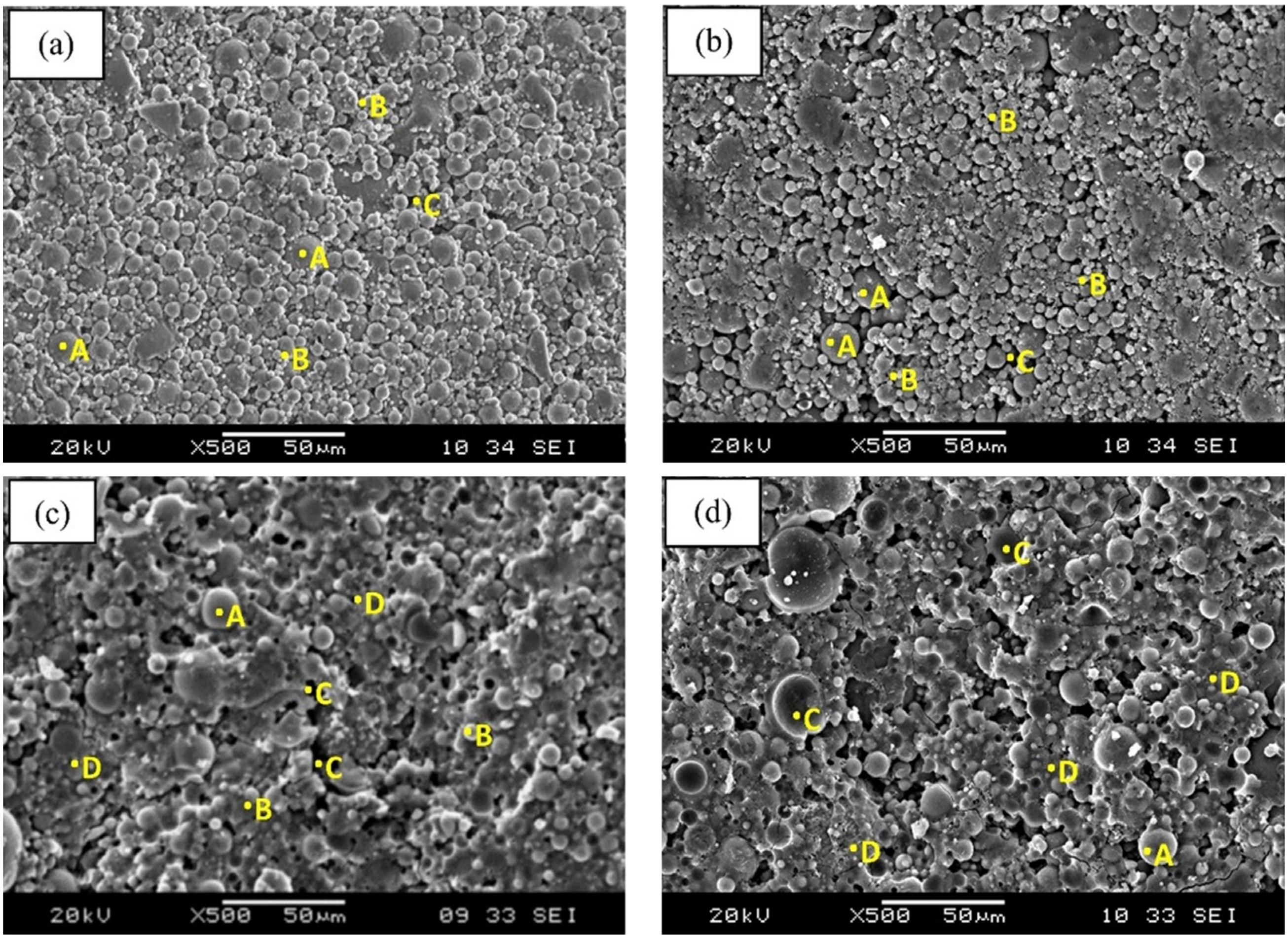


Fig. 4. Compressive strength of cold-pressed geopolymer with different NaOH concentration after 7 and 28 days

opment is limited at 28 days compared to that of 14 M NaOH concentration (109.6 MPa). The declined in compressive strength at 16 M NaOH concentration might be caused by the generation of geopolymeric matrix at a very early age which obstructs the reaction between aluminosilicate material and alkali activator at the later age [9].

3.3. Microstructural analysis

Fig. 5 shows the morphology of cold-pressed geopolymer with different NaOH concentrations after 28 days. A significant amount of unreacted fly ash and limited geopolymeric gel was observed in geopolymer that prepared from 10 and 12 M of



A: Inert fly ash; B: Globular unit; C: Pore; D: Geopolymer gel

Fig. 5. SEM micrographs of cold-pressed geopolymers with NaOH concentration of (a) 10 M; (b) 12 M; (c) 14 M; and (d) 16 M

NaOH solution. As stated in section 3.2, the low pH condition debilitates the chemical reaction between precursor and alkali activator which hindered the synthesis of geopolymer matrix. This consequently levelled off the compressive strength of geopolymer (Fig. 4). A lower degree of geopolymerization reaction was also noted by Chen et al. [6] where a remarkable amount of inert fly ash particles and pores were existed in the microstructure of geopolymer with 10 M NaOH concentration.

When the NaOH concentration went higher than 12 M, aluminosilicate gel formed and covered on the surface of inert fly ash. The geopolymer gel filled in the pores, and improved the compactness of geopolymer structure. The observation could be supported by the reduced percentage of porosity (Fig. 2). In comparison with geopolymer prepared by 14 M NaOH solution, geopolymer with 16 M NaOH concentration showed a less dense morphology. That is to say, the microstructure of geopolymer can be enhanced by increasing NaOH concentration, but it has an optimum range.

4. Conclusion

This experimental study depicted a change of mechanical achievement of fly ash based cold-pressed geopolymer with varied NaOH concentration. The highest 7-day compressive strength of 91.8 MPa can be achieved at NaOH concentration of 16 M, but the strength development was restricted at 28 days. The 28-day compressive strength (109.6 MPa) tended to be maximized using 14 M of NaOH concentration. Besides, the geopolymer specimen synthesized by 14 M NaOH concentration has the minimum percentage of porosity. The SEM results proved that geopolymer prepared from 14 M NaOH has the highest amount of sodium aluminosilicate gel. The geopolymer morphology is correlated to the result of compressive strength. By optimizing the NaOH concentration, cold-pressing method offer an alternative approach to fabricate a high strength geopolymer for applications as bricks, tiles and precast concrete.

This study has provided a basis understanding of the effect of NaOH concentration on the behaviour of cold-pressed geopolymer. However, other design variables such as solid-to-liquid ratio, Na_2SiO_3/NaOH ratio and pressing force also play a crucial role. Therefore, the study on the impact of these design variables could be the potential areas of future research. Moreover, the lack of research on the durability (chemical attacks, freeze-thaw cycles and efflorescence resistance) of cold-pressed geopolymer presents uncertainty for it to be used for practical applications. More investigations should be done to fill these gaps.

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