Geopolymer via Pressing Method: Aluminosilicates/Alkaline Solution Ratio as the Determining Factor

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Abstract. Aluminosilicates/alkaline solution (FA/AA) ratio has a great influence on the morphology and mechanical properties of geopolymer fabricated via pressing method. The aim of this paper is to evaluate the effect of FA/AA ratio on the performance of pressed geopolymer. The geopolymer mixtures were prepared at FA/AA ratio of 4.5-7.0 and compacted with uniaxial hydraulic press. The resultant specimens were cured at room temperature (30°C) for 7 and 28 days. The physical properties of the specimens were measured by porosity and water absorption analysis. The microstructure and strength of pressed geopolymers were determined. The results revealed that the pressed geopolymer with FA/AA ratio of 5.5 had the lowest porosity and water absorption. Furthermore, SEM micrographs proved that FA/AA ratio of 5.5 yielded the formation of well-compacted structure. Maximum compressive strength of 78.54 MPa was achieved.

1. Introduction

In 1970, Davidovits invented alkali-activated cements due to various catastrophic fires in France. These materials named “geopolymer” referred to the amorphous to semi-crystalline 3D network of silico-aluminate structures. The synthesis of geopolymer is an integrated process known as “geopolymerization” involves alkali activation of solid aluminosilicate materials by using alkali activator solution (metal hydroxide and/or metal silicate) [1, 2]. A vast variety of industrial waste can be employed in the production of geopolymer such as fly ash, slag and waste glass. Thus, geopolymers have the capability to solve the environmental problem regarding to the waste disposal and storage problems.

Normally, geopolymer is formed from casting geopolymer slurry in mould using a significant amount of alkali solution. This circumstance increases the possibility of forming geopolymer with high porosity. The existence of inhomogeneous and large pores acts as stress concentration point that prompts to mechanical failure. In this context, the application of pressure while reducing the liquid volume in geopolymer matrix might be the route in resolving the aforementioned problems [3-5].

In this study, by utilization of pressing method, solid aluminosilicates/alkaline solution (FA/AA) ratio plays an important role in governing the mechanical behavior of pressed geopolymer. However, there is limited number of related study have been reported especially in pressing method. Hence, it is
essential to evaluate the effect of FA/AA ratio to produce high mechanical strength pressed geopolymer.

2. Experimental

2.1. Material

Class F fly ash was used as the raw material to synthesize pressed geopolymer. It was collected from Manjung Coal-fired Power Plant, Perak, Malaysia. The chemical composition of the fly ash was determined by X-ray fluorescence (XRF) analysis as enlisted in table 1. The major components of fly ash included SiO₂, CaO, Al₂O₃ and Fe₂O₃. The morphology of fly ash used in this study is illustrated in figure 1. It was clearly shown that the fly ash had spherical shape with smooth surface.

| Compound | SiO₂ | CaO | Al₂O₃ | Fe₂O₃ | SO₃ | K₂O | Others |
|----------|------|-----|-------|-------|-----|-----|--------|
| Mass (%) | 36.70 | 19.10 | 18.70 | 17.15 | 3.04 | 1.78 | 3.53    |

The mixture of sodium hydroxide (NaOH) and sodium silicate (Na₂SiO₃) solution was utilized as alkali activator. The Na₂SiO₃ solution had chemical composition of 30.1% SiO₂, 9.4% Na₂O and 60.5% of H₂O with SiO₂/Na₂O ratio of 3.20. The NaOH pellets used was caustic soda with purity of 99.0%.

2.2. Preparation of geopolymer

10 M of NaOH solution was obtained by dissolving NaOH pellets with distilled water 24 hours prior to use. The alkali activator was prepared by mixing NaOH and Na₂SiO₃ solution in the ratio of 1.0. The solution was mixed with fly ash at aluminosilicates/alkaline solution (FA/AA) ratio of 4.5, 5.5, 6.5 and 7.0 until a homogeneous mix was achieved. The mixture was then poured into a cylindrical stainless-steel mold and manually pressed at 5 tons for 2 minutes. The geopolymer green body was removed from the mould and wrapped with plastic film. Lastly, the specimens were kept at room temperature (30°C) for 7 and 28 days.
2.3. Testing and analysis
The physical analysis was carried out by performing bulk density measurement and water absorption test. The compressive strength of pressed geopolymer specimens was evaluated in accordance with ASTM C 109 using Instron Machine Series 5569 Mechanical Tester with loading rate of 1 mm/min. A minimum of 3 samples were tested for each parameter, the average value of compressive strength was calculated. Besides, the morphology of pressed geopolymers at 28 days was determined by Scanning Electron Microscopy (SEM) analysis.

3. Results and Discussions

3.1. Porosity Analysis
The percentage of porosity of pressed geopolymer with different FA/AA ratio at 7 and 28 days are given in figure 2. The percentages of porosity of resultant geopolymers are in the range of 9.76 % and 15.16 %. Generally, the porosity of pressed geopolymer reduced from ageing time of 7 to 28 days, corresponding to the high degree of geopolymerization which resulting in a less porous structure. Geopolymerization process involves simultaneous occurrence of dissolution and condensation of precursors. In the presence of alkali activator, the precursors release free SiO_4 and AlO_4 which polymerize into an amorphous gel. The polymeric gel continues to grow bigger resulting in 3D aluminosilicate networks of geopolymer. Hence, it is supposed that the pore filling effect of geopolymer increases as time passes [6].

It is observable that the pressed geopolymer achieved the least percentage of porosity when FA/AA ratio is 5.5. This is mainly because, with FA/AA ratio of 5.5, the matrix has sufficient amount of alkali activator and tends to develop optimum geopolymerization. As a result, the pores of geopolymers are refined, thereby densify the microstructure of resultant specimens and reduce the percentage of porosity.

Figure 2. Porosity of pressed geopolymer with different FA/AA ratio after ageing for 7 and 28 days.

3.2. Water Absorption Analysis
Figure 2 depicts the percentage of water absorption of pressed geopolymer synthesized with different FA/AA ratio. The trend of water absorption is congruent with the result of porosity as shown in figure 3. This observation proves that the percentage of water absorption is relatable to the porosity of...
geopolymer. This is reasonable since the decrement of porosity at exposed surface contributes to a reduction of water absorption [7].

The pressed geopolymer exhibited an increment in water absorption when FA/AA ratio increases from 5.5 to 7.0. This might pertain to the pressing characteristic of geopolymer mix. The surplus solid content tends to produce a relatively dry mix which decreases the mix’s workability and consequently reduces matrix compactness. Based on the study done by Ranjbar et al. [8], the inadequate amount of alkali activator hamper the formation of geopolymeric gel, thus, results in a less dense geopolymer structure.

![Figure 3. Percentage of water absorption of pressed geopolymer with different FA/AA ratio after ageing for 7 and 28 days.](image)

3.3. Compressive Strength Analysis

The compressive strength of pressed geopolymer after ageing for 7 and 28 days are shown in figure 4. The results indicate that the 7-day strength of resultant specimens varied between 11.99 and 55.74 MPa, while 28-day strength changes between 16.77 and 78.54 MPa. As seen from the compressive strength result, the geopolymer with FA/AA ratio of 5.5 has maximum strength of 55.74 and 78.54 MPa after 7 and 28 days, respectively. These results suggest that FA/AA ratio of 5.5 is beneficial in promoting the dissociation of fly ash and yield the formation of rigid structure, at the same time avoiding the overflow of charge during compaction process.

However, the compressive strength of pressed geopolymer impaired when FA/AA ratio decreases to 4.5, attributes to the formation of viscous mix that increases the difficulty of compaction process. Similar to the finding, Posi et al. [9] stated that the extra liquid content will lead to the formation of highly workable mix which is unsuitable for pressing method.

As the FA/AA ratio adopted is higher than the optimum (FA/AA=5.5), the dissolution process of fly ash is hindered, and diminishing the strength development of geopolymer. Besides, it can be figured out that the 28-day strength of geopolymer with FA/AA ratio of 6.5 and 7.0 showed a limited strength improvement as compare to their 7-day strength. The key factor that prompts to this situation are because of the lack of alkali activator content obstructs the continual dissolution and condensation process of geopolymer.
3.4. SEM Analysis

Figure 5 illustrates the SEM micrograph of pressed geopolymer with different FA/AA ratio after 28 days. Geopolymer that produced from FA/AA ratio of 5.5 has a highly compacted geopolymer structure, composing with a tremendous amount of globular units, the densification of geopolymer is caused by the aids of pressure which contributes to strong bonding. Not only that, the optimum reactivity of fly ash has been achieved with FA/AA ratio of 5.5, evidenced by the compressive strength measurement as in figure 4.

A less compact morphology is observed when FA/AA is 4.5. The less-dense structure appears to level off the mechanical performance of pressed geopolymer. When FA/AA ratio went beyond 5.5, a significant amount of inert fly ash is embedded in geopolymer matrix. The fly ash particles remained unreacted due to insufficient liquid content that hinder the dissolution process of fly ash in geopolymer system. Since the liquid content decreases with FA/AA ratio, this limits the contact between fly ash and activator solution, and subsequently slowing down the dissolution rate of fly ash. Hence, it is non-surprising that the amount of inert fly ash at FA/AA ratio of 7.0 is higher than that at 6.5. It is established that FA/AA ratio is utmost important in determining the morphological characteristics of geopolymer that fabricated via pressing method.
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**Figure 5.** SEM micrographs of pressed geopolymers with FA/AA ratio of (a) 4.5; (b) 5.5; (c) 6.5; and (d) 7.0 after ageing for 28 days.

4. **Conclusions**

This paper has presented the results of an experimental study that was carried out to investigate the effect of FA/AA ratio on the behaviour of pressed geopolymer. It can be concluded that FA/AA ratio is one of the important aspects in controlling the morphology and compressive strength of geopolymer. When FA/AA ratio is equal to 5.5, a well-packed geopolymeric structure with the least porosity and water absorption formed. Besides, the highest compressive strength of pressed geopolymer could be achieved at FA/AA ratio of 5.5.

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