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Effect of Curing Profile on Kaolin-based Geopolymers

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Abstract

Depending on the processing conditions, geopolymers can exhibit a wide variety of properties and characteristics. Curing profile serves as a crucial parameter in synthesis of geopolymers. In this paper, the influence of curing temperature and curing time on the properties of kaolin-based geopolymer was studied. The samples were separated into several curing conditions; including curing at ambient temperature, 40°C, 60°C, 80°C and 100°C for 1 day, and up to 3 days. The compressive strength and SEM analysis of geopolymer products were evaluated. Results showed that curing condition has a significant effect on the mechanical properties of kaolin-based geopolymer. Generally, curing at ambient temperature was not feasible, while increase in temperature favored the strength development. In addition, prolonged curing time improved the geopolymerization process, and led to higher strength gain. However, curing at high temperature for a long period of time caused failure of the sample at a later age.

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Keywords: Geopolymer; Curing Temperature; Curing Time; Kaolin; Curing Profile

1. Introduction

Many parameters, such as the water content, the kaolinite ratio, and the type of metal silicate used, have a substantial effect on the final properties of the geopolymer. In particular, current work has shown

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that the thermal history of the source materials, such as kaolinite, as well as the curing regime for the geopolymer are important factors that must be taken into consideration when designing a kaolin-based geopolymer product for a specific application [1]. In the study of Alonso and Palomo [2], rate of polymer formation is influenced by parameters such as curing temperature, alkali concentration, initial solid content, etc. When the activator concentration increases, a delay in polymer formation is produced, whereas temperature accelerates its formation.

Effect of curing temperature on the development of the hard structure of metakaolin-based geopolymer was studied by Rovnanik [3], which aimed to analyze the effect of curing temperature (10, 20, 40, 60 and 80°C) and time on the compressive and flexural strengths, pore distribution, and microstructure of alkali-activated metakaolin material. The results showed that the treatment of fresh mixture at elevated temperatures accelerates the strength development, but in 28 days the mechanical properties deteriorated in comparison with results obtained for mixtures that were treated at an ambient or slightly decreased temperature. Other research also has reported that extra water decreased the compressive strength, whereas increased curing time or curing temperature increased the compressive strength [4]. Therefore, compressive strength depends on curing time and curing temperature, and water will give effect to the strength of the final product. In the study of Van Jaarsveld, Van Deventer and Lukey [1] towards the effect of composition and temperature on the properties of fly ash and kaolinite-based geopolymers, they showed that the initial curing at higher temperatures does not increase compressive strength substantially above that achieved by curing at room temperature. Curing at higher temperatures for more than a couple of hours does seem to positively affect the development of compressive strength. Curing at 70°C seems to substantially improve compressive strength compared to curing at 30°C for the same period of time. It is of interest to note that curing for longer periods of time at elevated temperature appears to weaken the structure, suggesting that small amounts of structural water need to be retained in order to reduce cracking and maintain structural integrity. Also prolonged curing at elevated temperatures breaks down the gelular structure of the geopolymer synthesis mixture, resulting in dehydration and excessive shrinkage as the gel contracts without transformed to a more semi-crystalline form [5].

Development of kaolin geopolymer in terms of their curing regime has not yet been studied. Therefore, in this paper, important parameters, such as curing temperature and curing time, were discussed based on the kaolin geopolymer.

2. Experimental Methods

2.1. Materials

Sodium Hydroxide (NaOH) powder used was classified as caustic soda micropearls, 99% purity with brand name of Formosoda-P, made in Taiwan. A technical grade sodium silicate solution (Na_2SiO_3) was supplied by South Pacific Chemicals Industries Sdn. Bhd. (SPCI), Malaysia. The chemical composition was comprised of 30.1% SiO_2 , 9.4% Na_2O and 60.5% H_2O with modulus $\text{SiO}_2/\text{Na}_2\text{O}$ of 3.2, specific gravity at 20°C = 0.0014 g/cm^3 and viscosity at 20°C = 0.4 Pa·s. Kaolin was supplied by Associated Kaolin Industries Sdn. Bhd., Malaysia. The general chemical composition is tabulated in Table 1, which was obtained from supplier's datasheet. The physical form of kaolin used was of powder type, and has minimum 40%, of particle size less than 2 μm and maximum 2% of moisture content. It was used as Si-Al cementitious materials. Distilled water was used throughout.

Table 1: Chemical Composition of Kaolin

| Chemical | SiO_2 | Al_2O_3 | Fe_2O_3 | TiO_2 | CaO | K_2O | Na_2O | MgO |
|----------|----------------|-------------------------|-------------------------|----------------|-----|----------------------|-----------------------|-----|
|----------|----------------|-------------------------|-------------------------|----------------|-----|----------------------|-----------------------|-----|

| | | | | | | | | |
|--------|---------------|---------------|-------------|-------------|-------|-------------|-------------|-------------|
| Wt (%) | 50.00 - 52.00 | 33.00 - 35.00 | 0.60 - 1.00 | 0.50 - 0.90 | <0.05 | 1.50 - 2.00 | 0.01 - 0.05 | 0.30 - 0.70 |
|--------|---------------|---------------|-------------|-------------|-------|-------------|-------------|-------------|

2.2. Sample Preparation

NaOH pellets and water were first prepared and mixed in a volumetric flask to obtain a concentration of 8M and then allowed to cool down to room temperature. NaOH solution was mixed with Na_2SiO_3 with $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio of 0.32 to prepare a liquid alkali activator, 24 h prior to use. Kaolin powder and alkali activator were mixed with the solid to liquid (S/L) ratio of 1.0, and stirred well for a few minutes by using mechanical mixer. The fresh cement paste was then rapidly poured into a 50 x 50 x 50 mm steel mould and the samples were compacted to approximately one half of the depth (about 1 in. or [25 mm]) of the mould in the entire cube compartment, and the paste was tamped in each cube compartment at each layer as described in ASTM C109 [6]. Finally, the samples were separated into 2 sets of curing conditions, one set was cured at ambient temperature, and the other set was cured in an oven at temperature of 40, 60, 80 and 100°C. The sample was cured for 1 day, 2 days and 3 days. Samples were sealed with thin plastic at the exposed portion of the mould during the curing stage.

2.3. Testing and Analysis Methods

Compressive strength tests of all specimens were evaluated according to ASTM C109/C109M-08 by using the Instron machine series 5569 Mechanical Tester. All specimens of different curing conditions were taken out of oven after 1 day, 2 days and 3 days of curing, and were put in room temperature until the day of testing. The compressive test was carried out to evaluate the strength development for the specimens. The samples were compressed at 1, 3, 7, and 28 days.

JSM-6460LA model Scanning Electron Microscope (JEOL) was performed to reveal the microstructure of kaolin geopolymers at different curing conditions. The specimens were prepared in powder form and were coated by platinum using an Auto Fine Coater; model JEOL JFC 1600, prior to examination.

3. Results and Discussion

3.1. Compressive Strength

The low reactivity of kaolinite needed sufficient time for the geopolymerization process to occur, and hence, required a certain period of time in order to increase the extent of geopolymerization for strength gain. Reduction in reaction time can be achieved when calcined raw materials such as fly ash, or metakaolin were used for geopolymerization compared to non-calcined materials such as kaolinite. At ambient temperature, the reaction of kaolinite is extremely slow as shown in **Fig. 1**, during the first 3 days of curing; the sample did not harden and only showed results at 7 days of ageing, with a very low strength gain. This result is supported by the work of Kirschner et al. [7], stating that ambient temperature was unfeasible due to a delayed in the beginning of setting.

From **Fig. 1c**, it could be observed that the sample cured at 80°C was reduced in strength at the ageing of 28 days when it is heat-treated for 3 days; whereas, if cured for 2 days, the strength at 28 days reported was quite similar to the strength tested at 7 days. This indicated that the strength cannot be further improved upon. A similar situation occurred to samples cured at 100°C for 2 and 3 days. When samples were cured at 100°C for 2 days, the strength decreased at ageing of 28 days; whereas, for samples cured for 3 days, the strength dropped by 7 days of ageing. Although the temperature of 80°C and 100°C initially led a faster strength gain compared to other curing temperatures, then strength dropped after few

days of ageing. This was probably because the compressive strength decreased on curing at a higher temperature for a longer period of time, as prolonged curing at higher temperature caused the occurrence of gel contraction, which results in loss of water molecules and shrinkage may occur, the granular structure of geopolymer mixture may break, and the samples may not transform to a more semi-crystalline form [5]. On the other hand, curing at a temperature of 40°C showed a slightly faster strength gain compared to curing at ambient temperature. Curing at 40°C and 60°C were similar in that the strength gained without dropping at longer ageing time, at least up to 28 days. It is estimated that curing a sample at 40°C and 60°C will permit a continuing gain in strength even over a longer period of time. However, curing at temperature of 60°C seems to be the best for kaolin geopolymers because it gained a faster initial setting compared to 40°C, and also led in strength when cured for 3 days. Increasing temperature favored the dissolution of reactive species, and contributed to higher strength gain. This finding was similar to previous research by Swanepoel and Strydom [8], where they investigated the utilization of fly ash and kaolinite clay in the geopolymeric material, and their data showed that compressive strength after 7 and 28 days was highest for the sample heated at 60°C for 48 h, which was cured at not very high temperature, and not very long period of time. In other words, high temperature with short curing time favored compressive strength development; low temperature required longer curing times in order to generate a significant increase in strength.

Therefore, it could be concluded that heat is beneficial for the strength development and curing at a temperature below 100°C has more significant contribution to the geopolymeric reactions in kaolin. The geopolymer will experience a substantial loss of moisture if the curing temperature is high, this is because small amount of water is needed to be retained in order to eliminate cracking and maintain structural integrity [9]. Prolonged curing time improved the geopolymerization process resulting in higher compressive strength but curing at higher temperature for longer period of time will distorted the reaction leading to failure of the sample on later age. In brief, compressive strength increased with the moderate elevation of reaction temperature. Kaolin geopolymer needed thermal activation in order to obtain good compressive strength properties.

3.2. Scanning Electron Microscope (SEM)

Fig. 2 shows the microstructure of samples with different curing profiles. Two temperature profiles at 60°C for 3 days and 80°C for 1 day showed the desired curing temperature and time where the compressive strength was gained day by day, indicating that geopolymerization reaction has occurred continually without deterioration at later age. From both microstructures, ageing from day 1 up to 28 days, the samples appeared to have more geopolymeric gel, structure of this paste experienced more growth and became denser and more compact which contributed to highest compressive strength at day 28. This structure proved that the compressive strength of the sample increased by prolonged ageing as shown in the results for compressive strength. Curing at 60°C, up to 3 days seems to be the best development result. On the other hand, as mentioned above, curing at higher temperature such as 100°C for longer period will cause the structure to have lower strength at later age (**Fig. 2g and 2h**). Values slightly decreased with rising temperature made the hardened structure loosely pack, less dense, hence less compact and had larger precipitates. This is shown by **Fig. 1b** where sample aged 28 days contributed to the decrease in strength compared to day 7 under the same curing regime. The rapid setting at higher temperature prevented the mixture from the formation of more compact and tough structure [3]. Decrease in strength observed at day 28 may be due to the heating in consequence which can cause partial water evaporation with formation of micro cavities [10]. Hollow cavities seen in the images were possibly due to the spaces left behind by dissolved kaolinite particle, where during the heating process, the geopolymer provided escape route for moisture without significantly damaging the geopolymer matrix [11].

According to a previous study [12], the rise in curing temperature led to the increase in the amount of unreacted materials, which can be seen in micrograph and this finding also reported by Qhatani Mohsen [13], with a loose grained structure with unreacted clays particles remained when processes at 150°C.

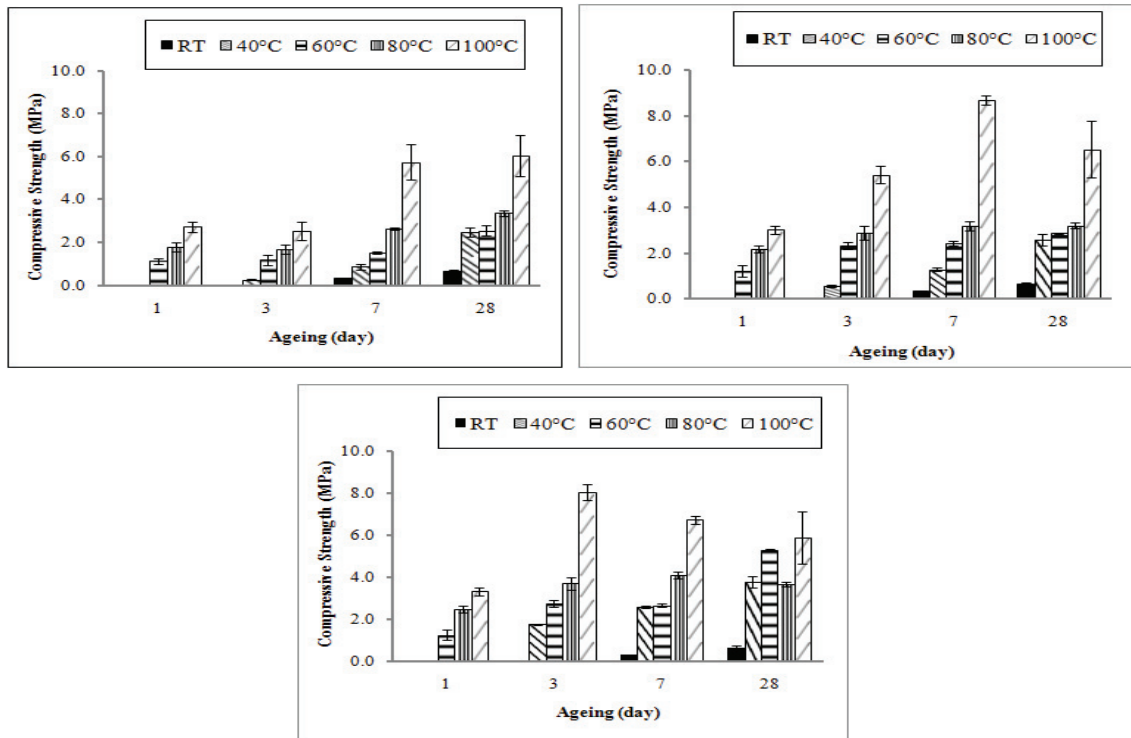


Fig 1: Compressive strength of kaolin geopolymers cured at different curing temperature for (a) 1 day; (b) 2 days; and (c) 3 days.

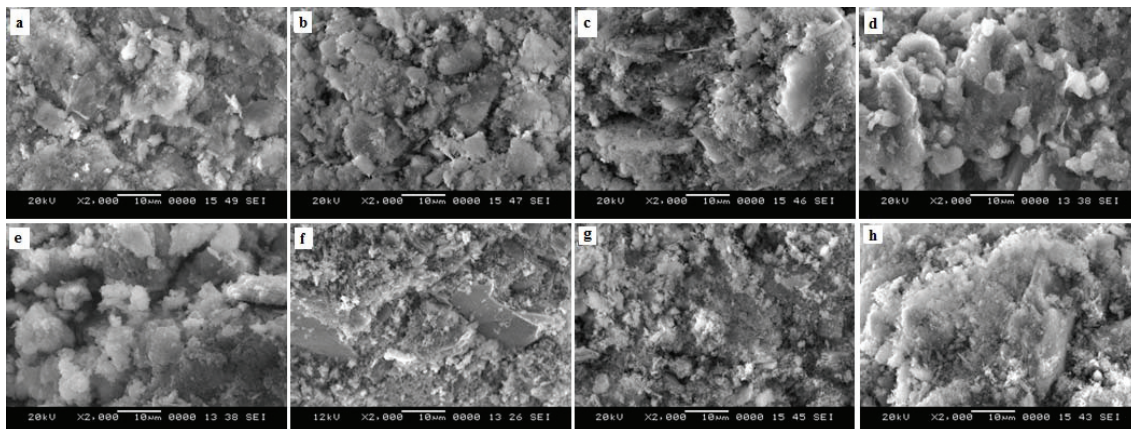


Fig. 4: SEM micrographs of geopolymers cured at 60°C for 3 days aged for 1 day (a), 7 days (b) and 28 days (c); geopolymers cured at 80°C for 1 day aged for 1 day (d), 7 days (e) and 28 days (f); and geopolymers cured at 100°C for 2 days aged for (g) 7 days, and (h) 28 days.

4. Conclusion

Geopolymers have emerged as novel engineering materials with the potential to form a substantial element of an environmentally sustainable construction and building products industry. Conclusions by this paper are as follows: (1) Curing at ambient temperature is unfeasible due to a delayed in the beginning of setting; (2) Increasing temperature favored the dissolution of reactive species and contributing higher strength gain; (3) Prolonged curing time improved the geopolymerization process; (4) Higher temperature curing for prolonged period distorted the reaction, causing partial water evaporation with formation of micro cavities which lead to sample failure at later age; and (5) Optimal development is achieved at 60°C up to 3 days supported by microstructure; the samples appeared to have more geopolymeric gel than other samples and experienced more growth and became denser and more compact.

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