

RECENT DISSERTATIONS ON KAOLIN BASED GEOPOLYMER MATERIALS

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Received: April 21, 2015

Abstract. Geopolymerisation also known as alkali activation of aluminosilicate occurs when mineral sources with high Al and Si content such as kaolin, blast furnace slag or fly ash are exposed to high alkaline environments, thus producing two to three dimensional Si-O-Al structure which exhibits both zeolitic and ceramic properties which is not typically present in traditional cement materials such as ordinary Portland Cement (OPC). Kaolin based geopolymers, inorganic polymers that are also alumino-silicate materials with impressive mechanical properties such as excellent coating material for fire resistant, poor electric conductor, chemically unreactive over a wide range of pH, non-abrasive, low energy consumption, poor permeability and emits less CO₂ which makes earth a better place. This paper is to review the progress made in kaolin based geopolymers that are currently in growing demand of researchers to replace existing eco-unfriendly materials.

1. INTRODUCTION

The demanding need of cement and concrete applications seeks ordinary Portland cement (OPC) alternate with greener final product which requires less energy and resource consumption. Geopolymers also known as 'alkali-activated aluminosilicates', 'hydroceramics', 'alkali-activated-cement', 'alkali-bounded-ceramics' and 'inorganic polymers' are the key solution to current issue [1-4]. Materials rich in Al₂O₃ and SiO₂ are aluminosilicate sources which is required for the exothermic geopolymerization reaction. Core Step of geopolymerization consist of three which are dissolution of Al and Si in alkaline

solution, diffusion of dissolved Al and Si ions to an inter-particle space, formation of agulated structure and lastly hardening of gel phase. Combination of sodium hydroxide (NaOH) and sodium silicate (Na₂SiO₃) are the most generally used alkaline solution for geopolymerisation process [5-9]. Sodium silicate acts as alkali activator, binder, plasticizer or dispersant while sodium hydroxide helps the dissolution of aluminosilicates sources. There are certain factors that influence the properties of geopolymers such as composition, type and relative amount of alkali activator, specific surface composition of source materials, and condition during the initial period of the geopolymerization process.

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Focus had been on the solid to liquid ratio (S/L ratio) and the alkaline activator ratio (Na_2SiO_3 -to- NaOH ratio) which have major impact on mechanical properties of geopolymers [10-16]. These crucial parameters strike the workability of geopolymer slurry. Previous work on fly ash geopolymer suggests that formulation ratio of raw source materials to alkaline activator by mass must be approximately 3 for geopolymerization process to occur [17].

Geopolymer is a new family of cementitious materials that is way more environmental friendly due to its less emission of CO_2 and energy consumption [1,18-20]. Geopolymer materials are generally low density due to the existences of micro sized and nano sized pores within the final products. Advantages of geopolymers as compared to ordinary portland cement (OPC) are high early strength, low permeability, less shrinkage, excellent durability, good fire and acid resistance. Commercial establishment and industrial application of geopolymer materials are still underrated due to lack of scientific literature availability [21]. Thus, it can be said that geopolymers are produced from the reaction of solid aluminosilicate precursors with alkaline solution at room or elevated temperature.

This predecessors may be of one type of mineral source or a combination of clays (usually kaolin, either raw or calcined kaolin), pozzolanics and various industrial by-products, such as slag and fly ash. Among these aluminosilicate sources, metakaolin or calcined kaolin has higher reactivity and relatively purer composition as compared to other materials [1,17]. Frequently used aluminosilicate sources are of kaolinite, fly ash, calcined kaolin, and chemically synthesized kaolin. Geopolymers are synthesized by polycondensation below 100°C at ambient pressure in an alkaline solution. In terms of past literatures, effects of calcined kaolin at high temperatures ($800\text{-}900^\circ\text{C}$) towards properties of post obtained geopolymer have not been elaborately discussed [22]. The nature and changes in geopolymer paste and the hardened final product are not fully explored, thus providing room for future works to meet the raising need of current various application as replacement of the typical cement binders. Not much attention was received by initial researchers that studied on how mineral composition and nature of kaolin and metakaolin effects the behaviour of paste and properties of hardened products [1, 23-25].

Kaolin, most versatile white mineral that possess outstanding properties such as chemically unreactive over a wide range of pH and good covering powder when used as a pigment or extender.

Secondary kaolins that are fine had been used as gloss materials due to their smaller particle size. Whereas platy kaolin particles, provides excellent coating opacity and printability [26]. Kaolin is also soft, non-abrasive, poor heat and electric conductor.

Particular applications of kaolin require very distinct specifications such as particle color, size, viscosity, and brightness whereas other uses requires none. These properties are important in paper coating industry, while in cement technology where the chemical composition is most crucial requirement. Kaolinite consists of silica tetrahedral sheets stacked alternately with alumina octahedral sheets and has the theoretical formula $(\text{OH})_8\text{Si}_4\text{Al}_4\text{O}_{10}$ and the theoretical composition 46.54% SiO_2 , 39.5% Al_2O_3 , 13.96% H_2O [27]. However usages of kaolin in terms of its suitability for ceramic based substrates, coating applications, cement and concrete industry is lacking.

Countable works have been done in the past, using calcined kaolin as coating materials for fire resistant applications [28-31]. These previous works explained and concluded that kaolin based geopolymer can withstand fire at high temperature, provided chemical compositions in the reaction system are altered accordingly.

Kaolin composes of kaolinite as its core mineral component with handful of secondary minerals, including anatase, quartz, dickite, halloysite and nacrite. In strong alkaline environments, dissolution rate and behaviour of quartz and kaolin differs as quartz has lower reactivity that kaolin. Structural characteristic of dehydrated halloysite, dickite, and nacrite differ while having similar chemical composition as kaolinite. Kaolinite has a sheet structure while halloysite has a tubular structure. In terms of hydroxyl groups, kaolinite has two out of the three hydroxyl groups contribute to the bond while, bonding between layers of dickite involves the collaboration of all inner-surface hydroxyl groups. It has been studied that various hydroxyl groups result in varying dehydroxylation behavior, which may impact the reactivity of kaolin. Thus, it can be concluded that these secondary minerals present in kaolin will affect the reaction process and final properties of geopolymer. Initial research on how secondary minerals content in kaolin affects the geopolymers should be investigated due to the various types and quantity of mineral composition in kaolin. This is important especially for the possible applications of geopolymers in future, which will involve huge amount of kaolin from multiple types of clay deposits. High bonding strength, low permeability and excellent

Table 1. Chemical composition of kaolin from various sources.

Chemical Composition of Kaolin	Kaolin de Beauvoir, France [32]	Associated Kaolin Industries, Malaysia [33]	Suzhou of Jiangsu, China [34]	South Pacific Industries, Malaysia [35]
SiO ₂	48.1	54.1	44.11	54
Al ₂ O ₃	36.9	33	37.21	31.7
Fe ₂ O ₃	0.26	4.41	0.55	4.89
TiO ₂	<0.25	1.34	0.31	1.41
CaO	<0.20	-	0.26	-
MgO	0.17	-	0.21	-
K ₂ O	1.9	5.67	0.58	6.05
Na ₂ O	<0.20	-	0.1	-
Li ₂ O	0.27	-	-	-
ZrO ₂	-	0.09	-	0.1
SO ₃	-	-	-	-
P ₂ O ₅	-	-	-	-
MnO ₂	-	0.11	-	0.11
Loss on ignition	-	1.28	15.5	1.74

thermal stability are some of the outstanding properties of kaolin-based geopolymers [1]. Thick gel is formed instantaneously upon mixing source material and the alkaline activator solution [17].

2. MATERIALS

Main raw material that was being studied is kaolin. Previous researches had obtained kaolin from various sources.

a) Raw commercial kaolin used was provided by kaolins de Beauvoir, France. This was the aluminosilicate source used for geopolymerization. Main composition of this raw kaolin was 81.5 wt.% of kaolinite, 3.5 wt.% of quartz, and 10 wt.% of micaeous minerals. The chemical composition of the raw kaolin is presented in Table 1 [32].

b) Fine particle with 40% of particle size less than 2 µm was purchased from Associated Kaolin Industries Sdn. Bhd, Malaysia. Raw kaolin was calcined at 800 °C for 2 hours. Table 1 tabulated the chemical composition of calcined kaolin [33].

c) Very fine kaolin from Suzhou of Jiangsu Province, China underwent thermal treatment at 600 °C, 700 °C, 800 °C, 900 °C, and 1000 °C for 6 hours. X-ray Fluorescence (XRF) was used to reveal the chemical compositions as tabulated in Table 1 [34].

d) South Pacific Industries Sdn Bhd, Malaysia supplied kaolin which was in powder form. The general chemical composition is tabulated in Table 1 which obtained from X-ray fluorescence (XRF) analysis [35].

3. METHOD

a) For the previous study on mechanical activation of kaolin based geopolymers, 8M NaOH solution was mixed with sodium silicate with ratio of 0.25 (sodium silicate/ NaOH solution) and left for 24 hours to obtain the alkaline solution for activation step. NaOH pellets was dissolved in deionized water and left to cool down to room temperature to obtain the NaOH solution. Kaolin undergoes dry milling at the speed of 250 rpm for various durations to be mechanically activated. Amount of kaolin used was 80 grams. After that, the kaolin powder was mixed into the activation alkaline solution with a mass ration of 0.26 (powder/solution) for 5 min at 125 rpm. A homogeneous paste was obtained. These was powdered and compacted at 125 MPa into a 20 mm steel mold. The as-obtained specimens were submitted to different curing conditions such as curing temperatures at 40 °C, 70 °C, and 100 °C, while the curing times were 24 hrs, 48 hrs, and 72 hrs. Each samples were analysed according to their ageing time which was (2, 7, 14, or 28 days) at room temperature [32].

b) As for the S/L ratio study, NaOH solution also with 8M was prepared at left at room temperature. Whereas, the alkaline solution was prepared by mixing sodium silicate and NaOH with a mass ration between 0.12 and 0.28 until transparent solution was obtained. This preparation was done 24 hours prior to use. Mechanical stirrer was used

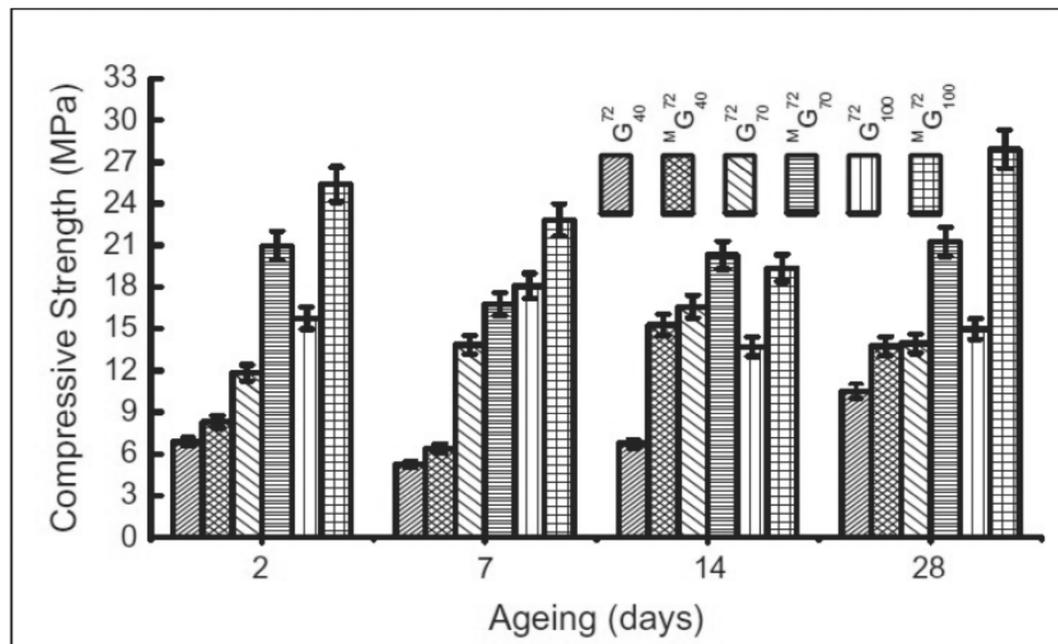


Fig. 1. Compressive strength of the different geopolymers versus ageing for the various curing temperature for 72 hours. reprinted with permission from D. H. Aji, G.L. Lecomte-Nana, G. Djétéli and P. Blanchart // Construc. Build. Mater. **42**(2013)105, (c) 2013 Elsevier.

to mix thermally treated kaolin with alkaline solution at a S/L ratio of 0.40-1.20 for couple of minutes to produce homogeneous slurry. Next, slurry was poured into a steel mold (50 mm x 50 mm x 50 mm), compacted according to ASTM C109 and sealed in films to prevent moisture loss. These samples were heated undisturbed in an oven for 3 hours at 80 °C. the solid geopolymers samples were then crushed using mortar and pestle, sieve through sieve mesh producing geopolymeric powder with desired 30–315 μm particle size. Geopolymer paste was obtained by adding powder with 22% of water. The geopolymer paste was used for various testing. Instron machine series 5569 Mechanical Tester was used to measure the compressive strength according to ASTM C 109/C 109M-08. Minimum three specimens were tested to evaluate compressive strength upon 7 days [33].

c) Role of water in Kaolin based geopolymers were also investigated by a previous researcher. Sodium hydroxide were mixed with sodium silicate and distilled water to prepare liquid activator, 24 hours prior to use. The modulus of sodium silicate used was 3.33, $\text{Na}_2\text{O}=9.98\%$, $\text{SiO}_2=27.10\%$). A constant S/L ratio was used throughout when calcined kaolin was mixed with alkaline activator, in order to study the effect of residual water on activity of calcined products. The obtained geopolymeric slurry was casted into cylinder plastic moulds to prepare specimens of $\text{Ø}25 \times 37.5$ mm. Various curing conditions such as in sealed bag, in air, in room temperature, humidity controlled room and elevated temperature were used in this work [34].

d) Influences of alkaline activator and S/L ratio on kaolin based geopolymers were investigated in an-

other study. For this work, 8 M NaOH solution was prepared in volumetric flask and left to cool down at room temperature. Na_2SiO_3 solution with $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio ranged from 0.16 to 0.36 was mixed with NaOH solution to prepare alkali activator solution 24 hours before use. The S/L ratio used to mix the kaolin and activation alkaline solution was from the range of 0.60 to 1.20. Mechanical stirrer was used to stir the mixture for couple of mins. Well mixed slurry was poured into 50 x 50 x 50 mm steel molds and compacted for about one half depth of mold and tamped in each cube compartment at each layer as shown in ASTM C109. Next, samples were left undisturbed in oven for 80 °C to be cured. Samples were sealed with plastic films during curing period [35].

4. RESULTS AND DISCUSSION

As per recent study on kaolin, mechanical activation of kaolin which is done by milling process influences the mechanical properties of kaolin based geopolymers. The results of compressive strength of raw kaolin and milled (mechanically activated) kaolin based geopolymers analysed after 72 hours of various curing temperatures are presented in Fig. 1.

Based on observation, in all cases mechanically activated kaolin based geopolymers exhibits (^MG) higher compressive strengths, comparatively to the raw kaolin based geopolymers (G). For curing time of 72 hours, increase of temperature up to 100 °C enhances the mechanical properties of the milled kaolin based geopolymers ($^M\text{G}_{100}$) and unmilled kaolin based geopolymers (G_{100}) after 28 days of age-

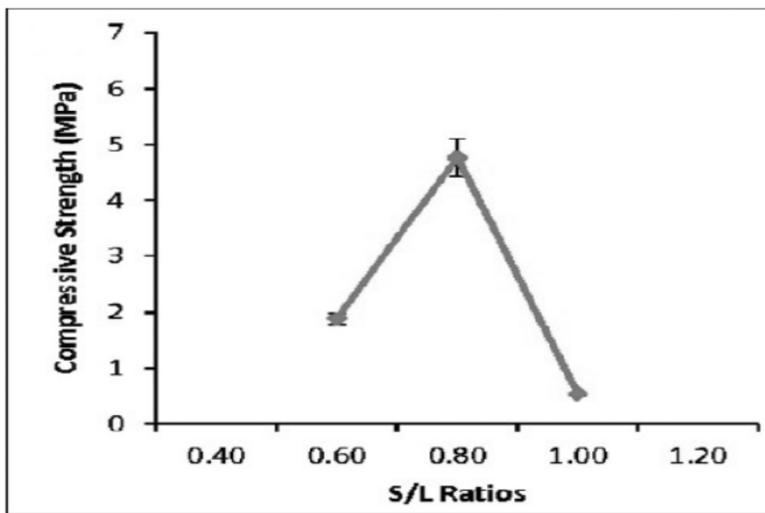


Fig. 2. Compressive strength at 7th day of resulted geopolymer pastes at various solids to-liquid ratio (constant $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio of 0.24). reprinted with permission from Y.M. Liew, H. Kamarudin, A.M. Mustafa Al Bakri, M. Bnhussain, M. Luqman, I. Khairul Nizar, C.M. Ruzaidi, and C.Y. Heah // *Construc. Build. Mater* **37**(2012) 440, (c) 2012 Elsevier.

ing. Mechanical activation of kaolin enhances the compressive strength to 51% for a curing temperature of 40°C , and of 86% for a curing temperature of 100°C , in comparison with the behavior of the raw kaolin based geopolymers after 28 days. In overview, the change of compressive strength with ageing time for a given curing temperature appears to be complex. In particular, a decrease of the compressive strength is observed at intermediate curing time at 7 or 14 days. It is noted that, after 7 days at low curing temperature (40°C) and 14 days at relatively high curing temperature (100°C), there is a significant decrease of the compressive strength for the milled and the unmilled raw based geopolymers, which increased subsequently after 28 days of curing. In addition, after 28 days of age-

ing, all geopolymers, except the one based on raw kaolin cured at 100°C , exhibit an increase in the compressive strength in comparison of that obtained after 2 days of ageing. Therefore, both mechanical activation and curing at 100°C improves the mechanical properties of kaolin based geopolymers. at higher temperature the mechanically activated kaolin shows greater mechanical properties as compared to the less sensitive raw kaolin geopolymers [32].

Geopolymer pastes with various S/L ratio (0.4, 0.6, 0.8, 1.0, and 1.2) were studied in a current recent work. Compressive strength of geopolymer pastes with varying S/L ratios at 7th day are shown in Fig. 2.

As observed, compressive strength peaked at S/L ratio of 0.80 but as the ratio was increased, it dropped gradually. For S/L ratio of 0.40 and 1.20, no proper compressive strength values were recorded because the mixes were extremely low viscous to allow for molding and were highly viscous for good compaction, respectively. The S/L ratio of 0.80 provided optimum workability and thus led to homogeneous slurry and optimal strength. The outstanding compressive strength makes geopolymer paste with S/L ratio of 0.80 with the optimum activator content that allows for dissolution of raw materials while not retarding the polycondensation rate during the geopolymer synthesis. Therefore, when water was added to the geopolymeric powder to produce resulted paste, the water content accelerates the polycondensation process due to the continual dissolution of residual raw materials and hydrolysis of generated Al_3^+ and Si_4^+ . Hence, it is observed that kaolin geopolymer paste with S/L ration of 0.80 provides the optimum compressive strength

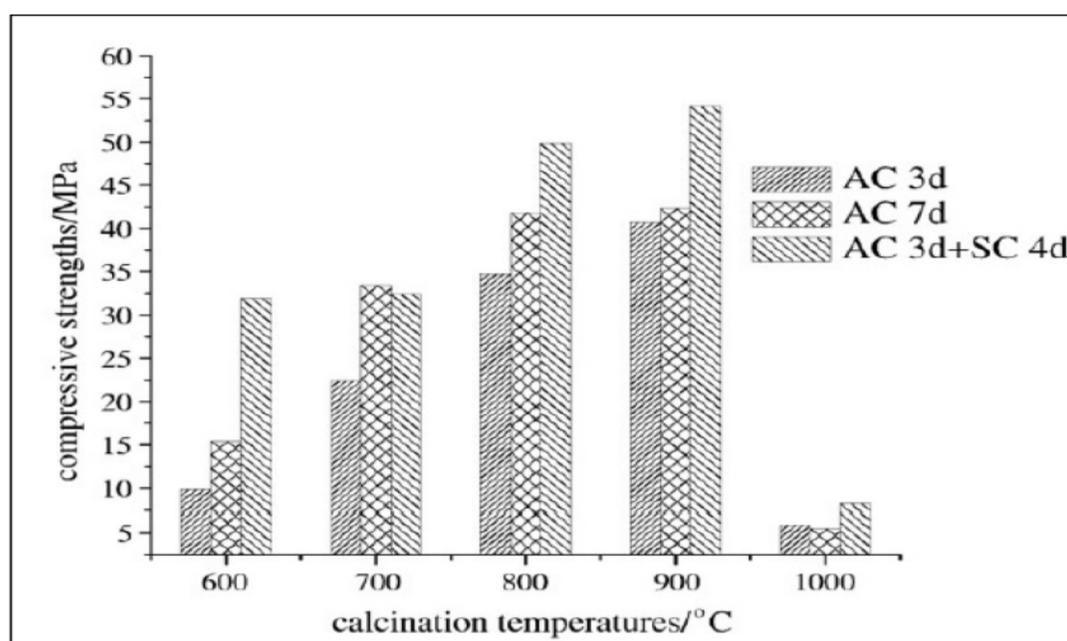


Fig. 3. Compressive strength development of different calcined kaolin based geopolymer. reprinted with permission from Z. Zuhua, Y. Xiao, Z. Huajun, and C. Yu // *Appl. Clay. Sci.* **43** (2009)218, © 2008 Elsevier.

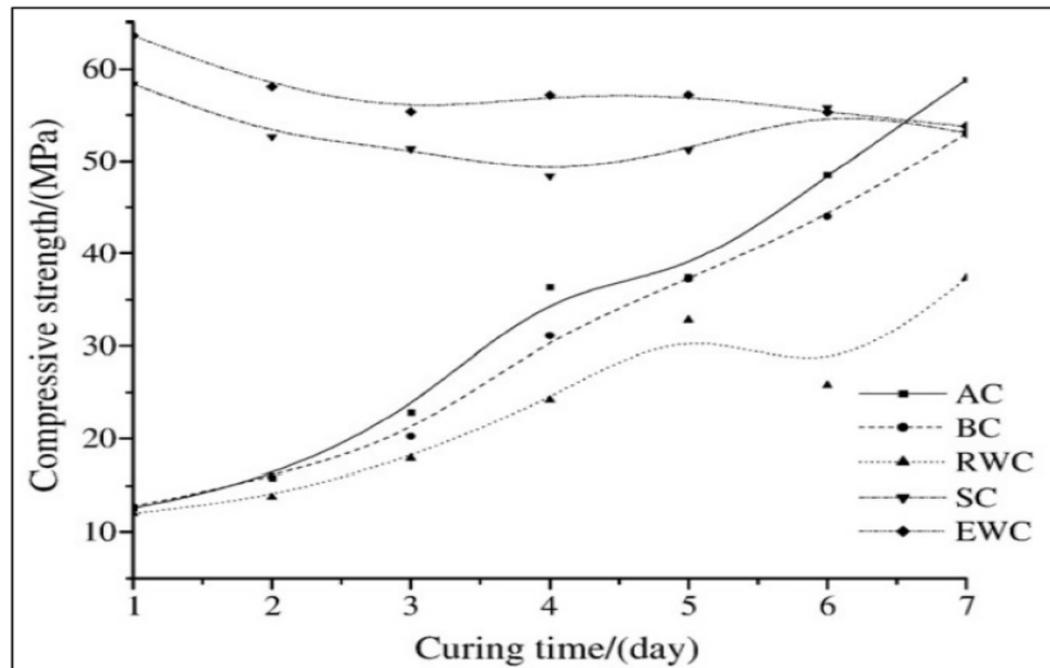


Fig. 4. Compressive strength development of geopolymers under different curing conditions such as air cured (AC), cured in high humidity (SC), in sealed bags (BC), immersed in room temperature (RWC) and immersed at elevated temperature (EWC). reprinted with permission from Z. Zuhua, Y. Xiao, Z. Huajun, and C. Yu // *Appl. Clay. Sci.* **43** (2009)218, © 2008 Elsevier.

when $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio is kept constant at 0.24 [33].

Fig. 3 shows the relationship between calcination temperature and the compressive strength of different calcined kaolin based geopolymers. It is observed that compressive strength significantly increases to a maximum at 900 °C and drops drastically at 1000 °C.

The compressive strength of generated geopolymers are dramatically different upon ageing for those cured in air (AC) and those cured in high humidity (SC). This is because the content of residual excess water in calcined kaolin differs. By thermogravimetry (TG) test (curve not shown), the mass fall at 700 °C, 800 °C, and 900 °C was 0.9%, 1.9%, and 2.3% compared to that at 600 °C, respectively. Thus, proving that change in mass is due to evaporation of structure water. With that, it can be assumed that activity of raw kaolin source material is affected by the residual water content. Excess water in source material that exist in form of hydroxyl is known to hinder workability of kaolin based geopolymers. It's difficult for structure water to be lost completely even at temperature higher than 950 °C [36]. TG test also shows that mass falls at 1000 °C by 2.7% compared to 600 °C, meaning the activity should have increased. The residual water reduces at 1000 °C (Fig. 3), compressive strength is observed to have decreased rapidly. This may be because at elevated temperature, more stable phases such as mullite and cristobalite are formed [37]. Hitherto, making residual water very important and crucial but not the only factor indicating the activity of calcined kaolin.

The compressive strength of geopolymers under different curing conditions are represented in Fig. 4. Those geopolymers that were immersed in water (SC and EWC) accelerates the geopolymerisation during the first 24 hours. At early stage, compressive strength of AC and BC geopolymers has increased largely when cured at 20 °C but after 7 days curing AC ones even reach 59 MPa with a little beyond that of 80 °C. Due to low temperature and excess water, geopolymers immersed in room temperature (RWC) showed lower compressive strength than that under the other four conditions. Excess water around the hydrolysis species, retards the polycondensation process. Also, when cured in water, reagent tend to leach out from surfaces of geopolymers, which contributes to the slow compressive strength development [34].

When a set of samples were cured in an oven at 80 °C, many visible cracks on these geopolymer surfaces (Fig. 5), which is due to dry shrinkage. Similar observation was obtained by another researcher on metakaolinite based geopolymers [38]. Thus, it can be assumed that crack-free geopolymer requires sufficient humidity during curing [34].

The effects of $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio on mechanical properties of kaolin based geopolymers were investigated. Figure 6 shows the compressive strength of kaolin geopolymers with various $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratios such as 0.16 (Mix 5), 0.20 (Mix 6), 0.24 (Mix 3), 0.28 (Mix 7), 0.32 (Mix 8), and 0.36 (Mix 9) at constant S/L ratio of 1.00. Sinusoidal fluctuated curve was obtained after 180 days. The compressive strength results fluctuated from day 1 to day 3 due to the various reaction rate and formation

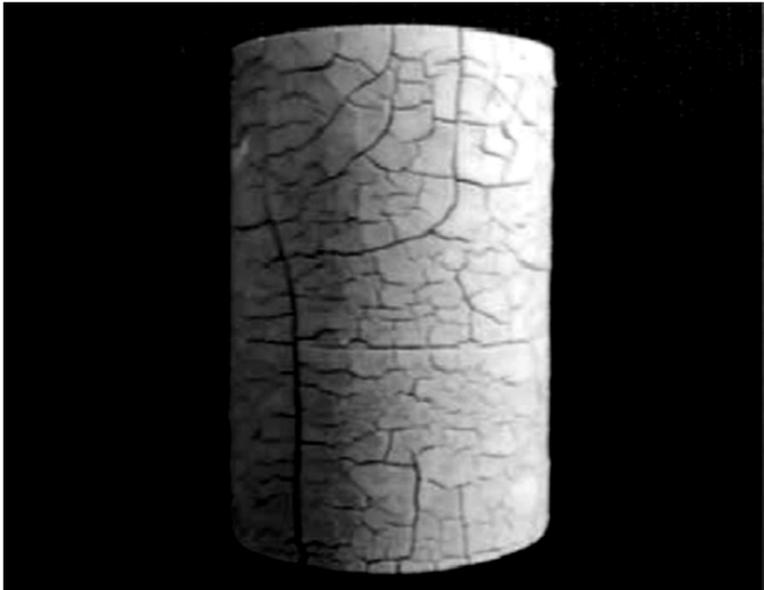


Fig. 5. Visible cracks on geopolymer surface after 80 °C curing for 1 day in an oven. reprinted with permission from Z. Zuhua, Y. Xiao, Z. Huajun, and C. Yu // *Appl. Clay. Sci.* **43** (2009)218, © 2008 Elsevier.

of the structure within the tested ratio as kaolinite has comparatively lower reactivity that requires sufficient time for interactions to occur. Content of Si and Al contributes to the reaction rate of gel [39]. Compressive strength can also help determine measured reactivity of kaolin.

Almost stable parabolic curve observed upon day 7. In general, compressive strength increased as the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio was increased until 0.32 (Mix 8). Ratio of $\text{Na}_2\text{SiO}_3/\text{NaOH}$ at 0.32 (Mix 8) appear to be the steadiest and have highest compressive strength increment rate as compared to other ratios. Sodium silicate, also known as waterglass liquid increases with increasing $\text{Na}_2\text{SiO}_3/$

NaOH ratio. This waterglass liquid was required for the geopolymerization process and acted as binder, alkali activator and dispersant or plasticizer [2]. Nevertheless, at $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio higher than 0.32, it is assumed that high amount of waterglass liquid hinders the geopolymerization process. Low reactivity of raw kaolin leads to slow dissolution of Al and Si ions into the system. Therefore, inadequate Al and Si ions were to be taken into reaction to form a rigid network of geopolymers. Geopolymer slurry was observed to become very sticky as the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio was increased thus providing high content of waterglass liquid. This stickiness was caused by the viscous nature of the waterglass liquid. These even worsen the condition where the the low reactivity kaolin with less dissolved Al and Si ions were unable to contact to each other. Thus, 0.32 was chosen as the best $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio for kaolin geopolymers.

5. CONCLUSIONS

- Mechanical activation of kaolin by dry ball milling improves the mechanical properties of kaolin based geopolymers at high curing temperatures.
- Solid to liquid ratio of geopolymer paste also plays an important roles in its compressive strength whereby S/L ratio 0.80 showed the optimum strength when $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio is kept constant at 0.24.
- Residual water in the form of hydroxyl is retards the activity of air cured kaolin based geopolymers to a significant temperature range. (until reaches 1000R°C).

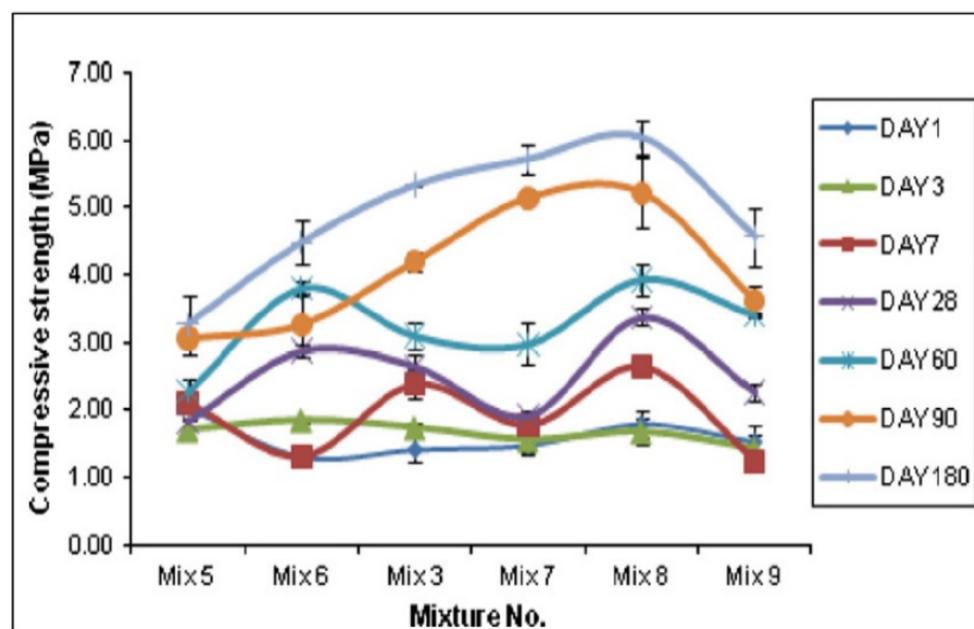


Fig. 6. Compressive strength of geopolymer samples with various $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratios (at constant S/L ratio of 1.00). reprinted with permission from C.Y. Heah, H. Kamarudin, A.M. Mustafa Al Bakri, M. Bnhussain, M. Luqman, I. Khairul Nizar, C.M. Ruzaidi, Y.M. Liew // *Construc. Build. Mater.* **35**(2012) 912. © 2012 Elsevier.

d) Sufficient humidity and water plays an important role in curing process of a crack free kaolin based geopolymer.

e) Kaolin based geopolymers that are cured under low temperature and excessive water will lower the compressive strength of geopolymers as polycondensation would be hindered and reagent will leach out from surfaces of geopolymers when cured in excess water.

f) It is believed that the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio chosen depended strongly on the workability of the mixes. Thus, 0.32 was chosen as the best $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio for kaolin based geopolymers with S/L ratio is at a constant 1.00.

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