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The Effect of Aggregate Particle Size on Formation of Geopolymeric Gel

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ABSTRACT

Due to enhanced mechanical and chemical properties, geopolymeric materials are a potential alternative to ordinary Portland cement and high-strength cement for the construction industry. The effects of aggregate particle size on the formation and the mechanical properties of geopolymeric materials are investigated in the current work. The addition of soluble silicates to the alkaline activator was found to increase the dissolution of silicate ions from the aggregate into the geopolymer binder phase. The results indicate that the surface area of aggregate influences the strength development of a geopolymer.

INTRODUCTION

The ability to re-use and add-value to industrial by-products such as fly ash and granulated blast furnace slag has become one of the major research fields in the last twenty years. Geopolymer technology has therefore emerged and is gaining international attention as it not only contributes towards recycling, and the minimisation of CO_2 emissions, but it also offers an economically feasible alternative to ordinary Portland Cement (OPC) or high strength cement (HSC). Davidovits in 1978 discovered a new inorganic aluminosilicate polymer, termed "geopolymer," that behaved similarly to zeolites and feldspathoids (Davidovits [1]). Geopolymers can be viewed as three-dimensional amorphous aluminosilicates. They are essentially formed by the dissolution and subsequent polymerisation of Al and Si precursors from an aluminosilicate source at a high pH (alkaline activation). Given the correct synthesis conditions, geopolymers can possess high early strength, low shrinkage, freeze-thaw resistance, corrosion resistance, high thermal stability, and acid resistance (Davidovits [1-2]).

It has been shown in the literature that the majority of research on geopolymer technology or alkaline cements can be summarised into the following categories:
(1) Characterisation and terminology of geopolymers (Davidovits [1-2]);
(2) Use of alkali-activated cementitious materials in the construction and building industry (Fernandez-Jimenez and Palomo [3] and Krivenko *et al.* [4]);
(3) Solidification and immobilisation of toxic metals using geopolymer technology (Van Jaarsveld *et al.* [5-6] and Palomo and Palacios [7]);
(4) Incorporation of different additives in geopolymers (Xu *et al.* [8] and Phair *et al.* [9-10]);
(5) Effect of different contaminants on the properties of geopolymers (Lee and Van Deventer [11-12] and Van Jaarsveld and Van Deventer [13]);
(6) Chloride diffusion and its relation to alkali-activated cement (Roy *et al.* [14]);
(7) Geopolymerisation of aluminosilicate minerals (Xu and Van Deventer [15-16]);
(8) Microstructural study of synthesised geopolymer (Xu and Van Deventer [17]);

(9) Roles of calcium in geopolymerisation (Yip and Van Deventer (18)).

Despite the extensive research effort on geopolymerisation, there is still a lack of mechanistic understanding of the geopolymerisation reaction. The actual formation and bonding mechanism at the interface of aggregates and gel is yet to be investigated. To fully understand the geopolymer process, a study on the formation of aluminosilicate gels and reactivity of solids in the gels is essential as it governs the mechanical and chemical properties of the resultant geopolymeric product.

Despite the attention given to the influence of the interfacial zone between aggregate and cement paste on the mechanical properties of concrete, Tasong *et al.* (19-20), there is currently no documented study on the reactivity of various solids (such as sand, basalt, and other aggregates) in geopolymeric gels. Properties including durability, bonding of the aggregate and matrix, density of the interfacial zone, and microstructural characteristics of the aggregate all need to be investigated prior to geopolymeric materials being accepted as possible alternatives to OPC or HSC. In the cement industry, it is often assumed that most rock aggregates are chemically inert and that their interaction with the cementitious matrix is to physically increase cement paste-aggregate bond strength. However, it is unknown at this stage whether a similar assumption on the role of aggregate inclusion can also be applied to geopolymeric materials.

In the present work, the effect of incorporating different sized sand, as the aggregate, in geopolymers was studied in an attempt to relate aggregate particle size and surface area to mechanical properties of geopolymeric gels. The role of sand in the geopolymeric gels was also investigated to relate the extent of reactivity of sand to formation of geopolymeric gels. Although the results of the current work are only preliminary, it has been found that reactivity of the sand aggregate in the geopolymer system has a significant effect on final material properties. This therefore provides scope for the use of additional aggregates, each possibly having a different behaviour in a geopolymer system, to design geopolymer concrete of exact desirable properties.

EXPERIMENTAL PROCEDURE

Materials

Metakaolin (MK) is highly reactive metastable clay that is essentially an anhydrous aluminosilicate obtained from calcining kaolin at a temperature range between 650-700°C (Newman (21)). Metakaolin used in the synthesis was obtained from ECC International, with a brand name of MetaStar 501. The sand was ground and sieved into three sized fractions; coarse (CS), medium (MS) and fine (FS). A magnet was used to minimise iron contamination during the grinding-sieving process. Sand was chosen as the aggregate source because its main composition is SiO_2, and due to its wide use in the cement and concrete industry. The composition of metakaolin and sand used were obtained by X-Ray Fluorescence (XRF) analysis, using a Siemens SRS 300 Instrument as shown in Table 1.

Table 1: Chemical composition of metakaolin (MK) and sand in mass %.

	SiO_2	Al_2O_3	CaO	MgO	Fe_2O_3	Na_2O	K_2O	TiO_2	MnO	P_2O_5	LOI
MK	52.22	41.41	0.08	0.26	0.49	0.01	1.73	0.01	0.01	0.13	1.66
Sand	99.27	0.16	0.00	0.00	0.00	0.16	0.01	0.07	0.00	0.05	0.28

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Table 2 shows the particle size distribution and surface area of metakaolin and each fraction of ground sand used in the synthesis. It is important to note that surface area of sand was calculated based on the assumption that sand has cubic geometry. Although not shown in the current work, SEM analysis was used to validate this assumption.

Table 2: Particle size distribution (microns) and surface area (cm^2/g) of metakaolin (MK), coarse sand (CS), medium sand (MS) and fine sand (FS).

	Surface area (cm^2/g)	Particle size distribution		
		d10	d50	d90
MK[a]	120000	-	2	10
CS[b]	4801	1425	1775	2375
MS[b]	12640	550	670	825
FS[b]	35120 [c]	184	275	394

[a]obtained from supplier. [b]obtained by sieving method using Endecotts Ltd, London, stainless steel sieve (BS 410/1986). [c]obtained by using Coulter Electronics Inc (Model LS 130).

The potassium silicate (K_2SiO_3) used was supplied by PQ Australia, Sydney (KASIL® 2236 (K32), weight ratio $SiO_2/K_2O = 2.23$, weight% $SiO_2 = 24.8$). Potassium hydroxide flakes (90-99% KOH) were obtained from Orica Australia. Distilled water was used throughout the experiments.

Synthesis

Pure metakaolin (MK) and coarse-, medium- and fine-metakaolin systems were prepared. Each sand fraction was dried in a laboratory oven at 40°C overnight prior to use. Potassium hydroxide of 7.05 M was prepared and cooled overnight to ambient temperature (System A). Alkaline silicate solution of potassium hydroxide and potassium silicate with a molar ratio of 1.2 (Ms = SiO_2/K_2O = 1.2) was mixed and cooled overnight (System B). The molar ratios were selected to maintain consistent water content available in geopolymer synthesis. The mass ratio between MK and the alkaline silicate solution was 0.8.

The mass ratio used between metakaolin and sand, MK/(MK+Sand), was 0.25. MK and each size fraction of sand were mixed for 3 minutes to obtain a homogeneous mixture. The alkaline silicate solution was subsequently stirred with the dry mix for 5 minutes until a uniform paste was formed. A mechanical mixer was used for stirring and mixing processes. The paste was poured into cylindrical moulds (50 mm diameter and 100 mm length) and allowed to cure for 24 h in a laboratory oven at 40°C, 53-57% RH. Samples were then extracted from the mould and hardened at room temperature until mechanical strength testing. All samples were cured at atmospheric pressure.

Analyses

Dissolution testing
The effect of particle size and surface area of aggregate on the leachability of silicate from metakaolin was determined by conducting dissolution tests containing MK and each size fraction of sand. The amount of alkaline activator added to the leaching matrix was based on mass ratio = 0.8, i.e. 12.5 g MK synthesised with 15.6 g of alkaline silicate solution. The alkaline activator in the leaching solution was diluted by a factor of 10. The same amount of alkaline activator solution was used to ensure

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constant water content and leaching condition that simulated real geopolymer synthesis. All leaching conditions are presented in Table 3. The slurries containing MK, sand and alkaline activator solution were stirred sufficiently to prevent sedimentation. At designated intervals: 1, 2, 4, 8 and 24 h, 10 ml of the suspension was collected and centrifuged at low speed for 10 minutes to separate the solid from the filtrate. Minisart membrane filter (0.2 μm) from Sartorius AG (Germany), was used to remove the remaining solids from the filtrates. The filtrate was further diluted by a factor of 12.5. Elemental composition of leached solutions was measured using ICP-OES (Perkin Elmer Optima 3000).

Table 3: Leaching conditions of various matrices and alkaline activator solutions in mass (g).

Matrix	MK	Sand				Alkaline Activator	
		Coarse	Medium	Fine	Total	KOH 7.05 M	KOH + K_2SiO_3
Basis	12.50		37.50		50.00		15.67
C1	50.00				50.00	156.70	
C2	12.50	37.50			50.00	156.70	
C3	12.50		37.50		50.00	156.70	
C4	12.50			37.50	50.00	156.70	
C5	50.00				50.00		156.70
C6	12.50	37.50			50.00		156.70
C7	12.50		37.50		50.00		156.70
C8	12.50			37.50	50.00		156.70
C9		50.00			50.00	156.70	
C10			50.00		50.00	156.70	
C11				50.00	50.00	156.70	
C12		50.00			50.00		156.70
C13			50.00		50.00		156.70
C14				50.00	50.00		156.70

Compressive strength testing
Compressive strength testing was performed according to Australian Standard (AS1012.9-1999) (22) using ELE compression machine at a pace rate of 1 kN/s. All values presented are the average of strength results of two samples. All samples were tested after 1, 7, and 28 days. Prior to strength testing, the top surface of the sample was capped with fast setting Boral Dental Plaster and left to dry at room temperature and atmospheric pressure for at least 3 h.

RESULTS AND DISCUSSION

Dissolution testing
It is believed that aggregates can influence ionic chemical interaction at the interface (Tasong *et al.* (19)). Different sized sand as aggregate is therefore anticipated to show different dissolution characteristics of Al-Si material. The leaching results obtained in the current work for matrices C1-C8 are presented in Figure 1. Results indicate the effect on addition of sand aggregate to MK in both 7.05M KOH (System A) and Ms= 1.2 KOH + K_2SiO_3 (System B).

It is shown in Figure 1A that for matrix C1 (MK only) the Si concentration in solution was substantially less than for matrices C2-C4. The shape of each dissolution curve was however very similar for matrices C1-C4, whereby for all matrices dissolution of Si for the first 8 hours was rapid, with only a minor amount of Si leaching thereafter

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(up to 24 h). As sand was added to the metakaolin system (matrices C2-C4), on average, the Si leached into solution increased by a factor of approximately 1.3 compared to C1 in system A (Figure 1a).
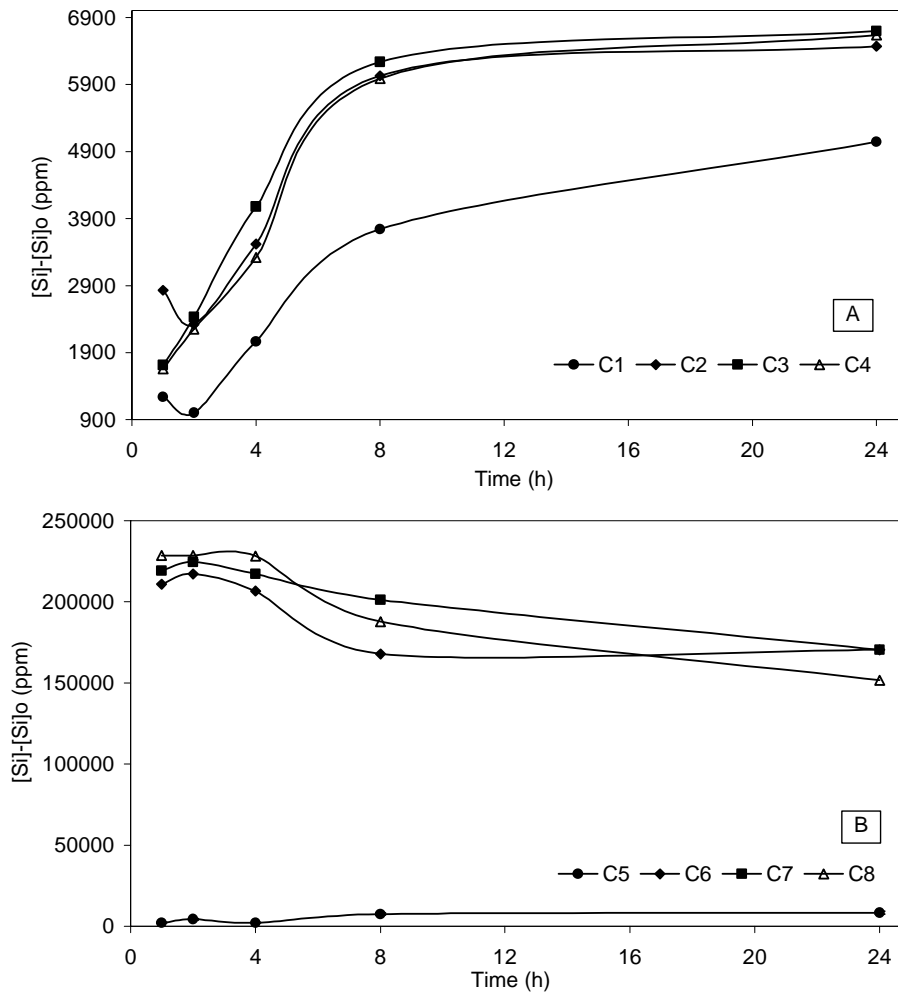
Figure 1: Silicon leaching characteristics (ppm) of (A) matrices C1 to C4 in 7.05M KOH and (B) matrices C5 to C8 in Ms=1.2 KOH + K_2SiO_3. [Si]= concentration of Si in the solution at time, t. $[Si]_0$= concentration of Si at t=0 (i.e. the added silicate).

At the initial leaching stage (t < 4h) it can be seen that C1 and C2 exhibit the same dissolution characteristics. It was found that the total Si concentration in solution decreased for the first 2 h, followed by an increase in Si concentration afterwards. In contrast, matrices C3 and C4 show an increase in Si dissolution up to 24 h. These slight differences in leaching characteristics are believed to be due to matrices C1-C2 having lower surface area available for ionic chemical interaction with alkaline solution than C3-C4.

Comparing system A with system B (Figure 1A and Figure 1B) it can be seen that the addition of soluble silicate to the leaching solution significantly increases the amount of silicon leached from the MK and sand. Figure 1b shows that the addition of soluble silicate, K_2SiO_3 to KOH with Ms=1.2, could induce significant changes to both MK and MK+sand dissolution characteristics as compared to the one without (System A).

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It is therefore believed that the addition of soluble silicates into the leaching solution has a positive effect on promoting significant structural breakdown of metakaolin (MK) and/or sand. The reasons for this phenomenon are unclear at this stage.

The resultant increase in Si leached from metakaolin in system B compared with system A (i.e. C1 and C5) was found to be by a factor of ~1.65. As in C1, C5 showed an increased dissolution of Si with time. It is important to note that matrices C1 and C5 contained 50 g of MK (no sand) in comparison to 12.5 g MK and 37.5 g sand for the other matrices studied (Table 3). It is also shown that more silicon is leached into solution after 24h in system B (C6-C8) compared with respective matrices in system A (C2-C4). This indicates that the addition of soluble silicates, as in real geopolymeric systems, substantially increases the extent of dissolution of Si from MK and sand. This result is consistent with the observation that the addition of soluble silicate into an alkaline activating solution enhances the dissolution of geopolymer precursors, which are necessary to initiate the geopolymerisation reaction (Xu and Van Deventer (15)).

The results presented in Figure 1 also show that differences in particle size and surface area of sand do not significantly influence the rate of silicon leached in either system A or system B, i.e. matrices C2-C4 or C6-C8 did not exhibit a significant difference in Si concentration in solution with time. However, it is observed from Figure 1B that the concentration of Si in solution increases initially, and then decreases over time to 24 h (matrices C6-C8). The decrease in silicon concentration after some time is caused by precipitation of aluminosilicates on the surfaces of the particles. An initially higher Si dissolution was observed for matrix C8, which indicates that sand having a larger surface area interacts to a greater extent with soluble silicates and MK to release a greater amount of Si into the leaching solution. This characteristic was also found in C6-C7, at which coarse sized sand addition to MK (Matrix C6) induced the lowest initial Si dissolution. It is proposed that larger surface area aggregate may promote chemical interactions between aggregate-MK and alkaline solution, thereby affecting the formation of geopolymeric gels. This is supported by the observation that a thin gel formed on the surface of the C8 leaching solution after 24 h. In system A, matrices C2-C4 did not show any formation of a gel on the leaching solution.

In order to investigate the effect of particle size and surface area of sand on the dissolution of silicon in both systems, each size fraction of sand (C9-C14) was leached without the addition of MK to the system (Table 3). Figure 2 presents the leaching results obtained for matrices C9-C14 in systems A and B. It is important to note that these matrices did not contain any metakaolin.

Comparing Figure 2A with Figure 1A, it is shown for system A (leaching with KOH) that only a minor amount of silicon was leached from the sand (C9-C11). In contrast, for system B (leaching using KOH and K_2SiO_3) it is shown that a significant amount of silicon leached from the sand into solution (C12-C14). Moreover, the amount of silicon in solution was approximately 13 times greater than that observed for matrix C5 (MK only). These results clearly indicate that the addition of potassium silicate during leaching has a significant effect on the dissolution of silicon from sand.

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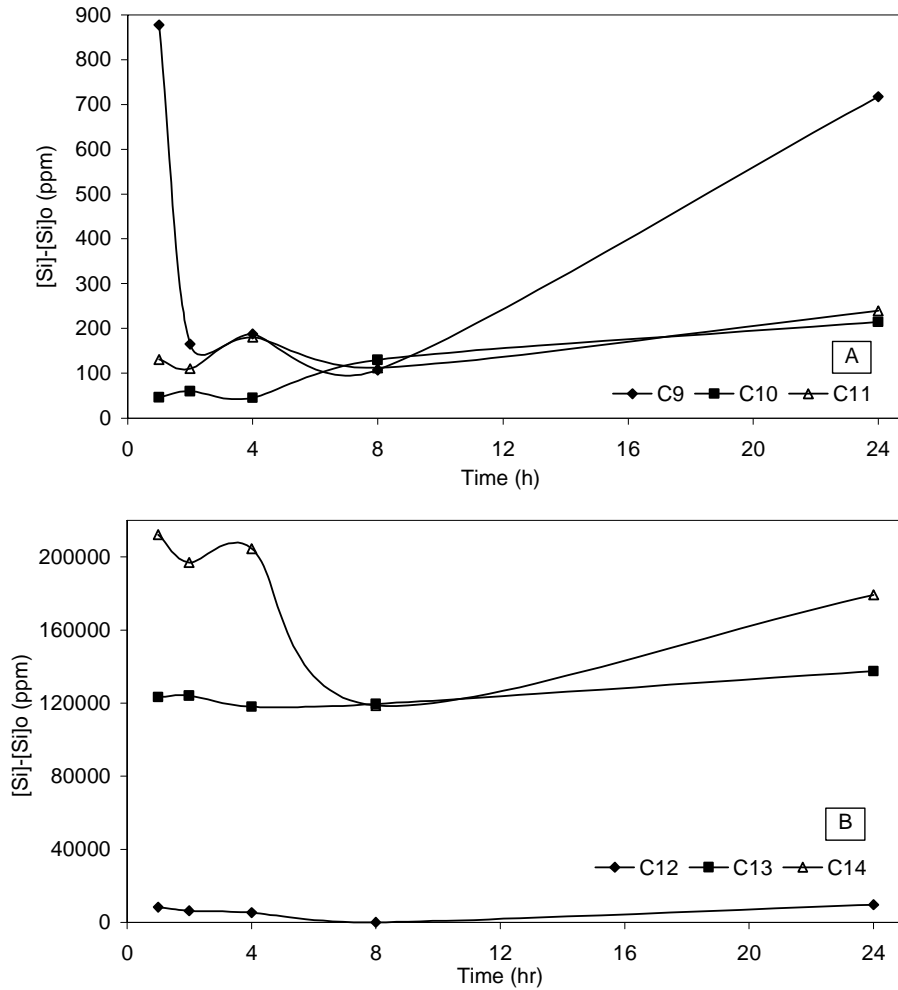
Figure 2: Silicon leaching characteristics (ppm) of (A) matrices C9 to C11 in 7.05M KOH and (B) matrices C12 to C14 in Ms=1.2 KOH + K_2SiO_3. [Si]= concentration of Si in the solution at time, t. $[Si]_0$= concentration of Si at t=0 (i.e. the added silicate).

It can also be gathered from Figure 2B that the dissolution of silicon from sand is affected by the particle size and surface area of sand particles. The coarse sand (C12) showed the least amount of leached silicon, followed by medium (C13) and then fine sand (C14). This is to be expected because the rate of dissolution of silicon should be greatest for the smallest particle size. It is proposed that the presence of large surface area sand contributed to early ionic chemical interaction with soluble silicates. These leaching tests conducted on each size fraction of sand for both systems A and B demonstrate that the sand aggregate is chemically active in a geopolymer system.

Compressive strength testing
Four types of geopolymer samples were produced (MK without sand, MK with coarse sand, MK with medium sand, and MK with fine sand) and their compressive strengths determined for different time periods. The alkaline activator used during synthesis was system B, Ms= 1.2, KOH + K_2SiO_3. The compressive strength of the binder containing different sizes of sand aggregates studied at 1, 7 and 28 days is presented in Table 4.

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Table 4: Compressive strengths of matrices using Ms= 1.2

Matrix	Al-Si Source	Sand	Compressive Strength (MPa)		
			1 d	7 d	28 d
A1	MK	Coarse	22.7	19.8	17.4
A2	MK	Medium	35.0	27.9	30.9
A3	MK	Fine	37.7	48.9	41.1
B1	MK	None	20.8	23.7	27.7

For the geopolymeric binder containing metakaolin and no sand, it was found that the strength of the matrix increases with time (i.e. 20.8 MPa after 1 day, 27.7 MPa after 28 days). As shown for samples A2 and A3, the inclusion of medium- and fine-sand had the effect of improving the overall strength of each sample compared to B1. For sample A1, it is shown that the 1 d strength was 22.7 MPa and that the strength of the sample decreased with time (at 28d, strength is 17.4 MPa). Although the reason for this is still unclear, it is believed that the coarse sand (CS) may not provide enough interfacial area for geopolymer paste to bind sufficiently; thereby resulting in a lower strength. In contrast, sample A2 showed a decreasing compressive strength between 1 to 7 days of curing, and strength gain afterwards.

It is suggested that the trend in compressive strengths for matrices A1-A3 and B1 can be related to the leaching characteristic exhibited by inclusion of each fraction of sand (Figure 1 and Figure 2). Although only a small difference in silicon dissolution was found for C6-C8 in dissolution testing, it was proposed that the ionic interaction of sand in geopolymerisation would contribute towards mechanical strengths of the resultant geopolymer. Referring to Table 4, it can be seen that an optimum sand size existed that gave the highest or stable compressive strength of the resultant matrix. As a general trend, sand with largest surface area, i.e. fine sand (FS) provided early strength, whereas sand with smallest surface area, i.e. CS was not able to provide sufficient interfacial area to bind with the geopolymer paste. This result has indicated that despite the increase in strength of sample by addition of sand to the matrix, each size fraction of sand has a different effect on the early-age and later-age strength of the resultant product.

It is proposed that dissolution of silicon from MK and sand (C1-C14) and optimum strength of the resultant geopolymer are associated with the binding area of aggregate. Table 2 lists the surface area of metakaolin and each size fraction of sand used in the present work. The surface area of FS was ~7 times of CS and ~3 times of MS. It is anticipated that the difference in surface area can affect the ingression of water into interfacial transition zone (ITZ). The role of water in a geopolymer system is believed to assist in ionic chemical interaction between the alkaline activator and the aluminosilicate source thus resulting in polymerisation and formation of geopolymeric gels. Hence, it is proposed that the presence and amount of water in the geopolymer system are related to the properties of the resultant geopolymer.

It is proposed that inclusion of low surface area aggregate, CS into the matrix (A1) resulted in less potential binding area to retain water required during the synthesis, and as a result, poor resultant geopolymer formed as shown by the low and decreasing strength trend. The effect of rapid hydration was also found to hinder the diffusion process in cement synthesis that eventually resulted in a lower compressive strength (Kim *et al.* (23)). As discussed earlier, these results indicate that there exists an optimum surface area for binding of aggregate to MK that will give stable strength development.

CONCLUSION

The addition of soluble silicates during leaching was found to increase significantly the dissolution of silicon from metakaolin and sand aggregate compared with a KOH solution only. Moreover, the addition of sand into the leach containing alkaline soluble silicate increased the structural breakdown of metakaolin, which resulted in the increased formation of silicate precursor species needed to form the aluminosilicate gel. Solution chemistry of KOH and K_2SiO_3 was found to affect the rate of silicon dissolution in the geopolymer system. Different sizes of sand aggregate were found to have an important role in the compressive strength of the resultant geopolymer matrix. Preliminary results have indicated that comparably higher and more stable strength values can be achieved when there is an optimum surface area for interfacial bonding between the geopolymer matrix and the aggregate. The rate of water loss from the interfacial bonding zone as well as the packing density of the aggregate have also been suggested to be important factors that contribute to geopolymer strength. The results of the current work enhance the understanding of aggregate effect on geopolymer synthesis and final compressive strength.

ACKNOWLEDGEMENT

NOTATION

C   Celsius
g   Gram
h   Hour
m   Metre
M   Molar
Ms   Modulus
N   Newton
RH   Relative Humidity
s   Second

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