Factors affecting the geopolymerization of alkali-feldspars

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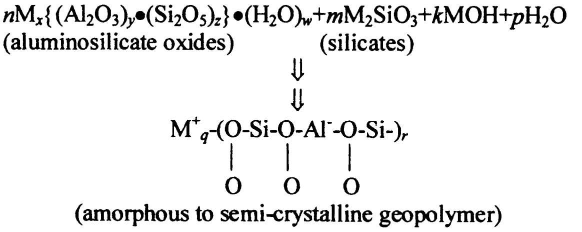
Abstract

Geopolymerization is a process by which Al-Si minerals undergo dissolution and polymerization in an alkaline solution to form a monolithic material with a high mechanical strength (Davidovits, 1991, 1994). In this study, the factors affecting the geopolymerization of Na-feldspar (albite) and K-feldspar (microcline) were investigated. It was found that both Na-feldspar and K-feldspar are reactive in geopolymerization with the resultant geopolymers showing a high compressive strength. The leaching ability, the proper molar ratio of Si/Al in the geopolymeric gel phase and the higher average hardness of the undissolved particles remaining in the geopolymers are found to greatly affect the ultimate compressive strength of the formed geopolymers.

Key words: Geopolymerization, Mineralogy, Albite, Microcline

Introduction

Invented by Davidovits in the late 1970s (Davidovits, 1991, 1994), geopolymers are amorphous to semicrystalline three-dimensional aluminosilicate polymers. Geopolymerization occurs in highly alkaline solution with aluminosilicate oxides and silicates (either solid or liquid) as the reactants. A geopolymeric reaction is described as



$$\begin{array}{c} n\mathrm{M}_x\{(\mathrm{Al_2O_3})_y\bullet(\mathrm{Si_2O_5})_z\}\bullet(\mathrm{H_2O})_w+m\mathrm{M_2SiO_3}+k\mathrm{MOH}+p\mathrm{H_2O} \\ \text{(aluminosilicate oxides)} \qquad \text{(silicates)} \\ \Downarrow \\ \Downarrow \\ \mathrm{M}^+_q\text{-}(\underset{\underset{\mathrm{O}}{|}}{\mathrm{O}}\text{-Si-}\underset{\underset{\mathrm{O}}{|}}{\mathrm{O}}\text{-Al}^-\text{-}\underset{\underset{\mathrm{O}}{|}}{\mathrm{O}}\text{-Si-})_r \\ \text{(amorphous to semi-crystalline geopolymer)} \end{array} \qquad (1)$$

where M = Na and/or K.

It is noted from Eq. (1) that theoretically any Al and Si containing mineral can serve as the source material in geopolymerization. More than 75% of the earth's shell consists of Al-Si minerals and silicate minerals (Dietrich and Skinner, 1979). Whether these naturally occurring minerals can be used in geopolymerization or not is important for the potential commercialization of this new technique. Xu and van Deventer (2000) investigated the ability of sixteen naturally occurring aluminosilicate minerals to undergo geopolymerization. Each of these minerals has a different crystal structure, chemical composition, density, hardness and degree of paragenesis. The results obtained by Xu and van Deventer (2000) indicate that natural Al-Si minerals with a framework structure have a higher reactivity than many other structures in geopolymerization, and KOH rather than NaOH solution produced a geopolymer with a higher compressive strength. However, it was also found that a higher content of K_2O in Al-Si minerals negatively affected the resultant geopolymers.

Alkali-feldspar is a large group of naturally occurring framework structured Al-Si minerals, which is also a group of common industrial minerals possessing homogeneous properties. The alkali-feldspars albite ($NaAlSi_3O_8$) and microcline ($KAlSi_3O_8$) have the same linked four-members ring structure. The chemical compositions of albite and microcline are very close to each other, except for the type of alkali metal. Thus, the industrial alkali-feldspars (Na-feldspar and K-feldspar) have been selected in the present work for the purposes of studying their abilities to undergo geopolymerization. Kaolinite, a clay mineral, has been added in geopolymerization as a source of Al_2O_3 during previous studies (Van Jaarsveld et al., 1998; Van Jaarsveld and van Deventer, 1999; Phair et al., 2000). To compare the geopolymeric systems studied here to previous work, kaolinite has been added as one of the reactants in the present work as well.

Experimental procedures

Materials. Kaolinite, grade HR1/F, was purchased from Commercial Minerals, Sydney, Australia. The kaolinite has a particle size of 50% less than 0.5 µm and 1% greater than 38 µm. Soda and potash feldspars were purchased from Minerals Corporation Ltd., Broken Hill, Australia. Both feldspars have a particle size of 100% less than 53 µm. The compositions of the kaolinite, Na-feldspar and K-feldspars were determined by X-ray fluorescence (XRF) analysis. The results, together with their average density and hardness (Nickel and Nichols, 1991), are presented in Table 1. Sodium silicate solution (Vitrosol N40) was supplied by PQ Australia and had a composition of 8.9% (by weight) Na_2O and 28.7% (by weight) SiO_2. Distilled water and analytical grade NaOH and KOH were used throughout all experiments.

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Table 1 — Mass composition, density and hardness of Na-feldspar, K-feldspar and kaolinite.

Element as oxide	Na-feldspar	K-feldspar	Kaolinite
SiO_2	70.9	67.1	54.4
Al_2O_3	17.0	17.6	29.4
Na_2O	9.75	3.16	0.2
K_2O	0.43	10.6	0.2
CaO	0.31	0.18	0.2
Fe_2O_3	0.07	0.21	1.4
MgO	0.04	0.04	0.2
MnO	<0.01	<0.01	–
P_2O_5	0.14	0.26	–
TiO_2	0.045	0.01	2.8
Loss on ignition	0.75	0.62	11
Density (g/cm^3)*	2.62	2.57	2.62
Hardness (Mohs.)*	6.5	6.5	2.3

*Density and hardness cited from Nickel and Nichols (1991).

Leaching tests and geopolymerization. A specified mass of M-feldspars and kaolinite was mixed with 20 mL MOH solution (where M = Na and/or K) at room temperature for 18 hours using a magnetic stirrer. After centrifuging, the clear liquid part of the solution was diluted to 0.1M alkaline concentration and neutralized by concentrated HCl to $pH < 1$. Because the Al and Si in diluted solutions were less than 50 and 100 ppm, respectively, no precipitation would occur (Lindsay, 1979). A Perkin-Elmer Optima 3000 ICP-OES was used to analyze the concentrations of silicon and aluminum leached in the solution. The leaching results of M-feldspars and kaolinite for three alkaline concentrations 2M, 5M and 7.5M are presented in Figs. 1 through 6.

A total of 950 grams of kaolinite and Na-feldspar and/or K-feldspar were mixed for ten minutes before an addition of 32 mL of sodium silicate and 245 mL of MOH solutions (where M = Na and/or K). The subsequent mixture was shaken by a FRITSCH vibratory shaker for another three minutes. The resulting paste was then transferred to a 50-mm-diam, 100-mm-high polyvinyl chloride (PVC) cylinder mould. The mould was left in an oven at 45°C for 24 hours for setting. After being removed from the mould, the sample was left in the oven for further setting and hardening at 45°C for 27 days. After this time, the compressive strength of each sample was tested using a Tinus Tolsen Compressive Strength testing machine. Two samples of each geopolymer mixture were tested, with the average compressive strength values depicted in Figs. 7 through 9.

X-ray diffraction and SEM/EDX analysis. X-ray powder diffraction (XRD) patterns (from 5 to 40 degrees (2θ)) were recorded on a Philips PW 1800 diffractometer using Cu K^{α} as a radiation source and scanning rate of two degrees per minute.

A JEOL JSM-840 microscope with Tracor Northern EDAX system was used for SEM/EDX analysis. The accelerating voltage of 20 kV was used throughout the SEM/EDX analysis. Samples for JSM-840 analysis were placed on the sample holder supported by carbon conductive paint followed by a one-minute sputter coating of gold.

Results and discussion

The dissolution of kaolinite, Na-feldspar and K-feldspar. Dissolution is the first step of geopolymerization. Normally, it is believed that a faster leaching would result in a faster gelation and setting with the resultant geopolymers possessing a high compressive strength (Xu and van Deventer, 2000). The investigation of natural Al-Si minerals shows that NaOH, compared with KOH, is able to leach more Al and Si (Xu and van Deventer, 2000; Xu et al., 2001). The concentrations of Al and Si complexes were found to increase significantly (Xu and van Deventer, 2000; Xu et al., 2001) when the concentration of MOH (where M = Na and/or K) increased from 2M to 5M and further to 10M.

Figures 1 and 2 depict the concentrations of Al and Si leached from kaolinite in NaOH and KOH solutions, respectively. The leached Al and Si complexes, as noted, increased greatly along with an increase in the concentrations of NaOH and KOH solutions, which is consistent with the general trend discovered from the study of other Al-Si minerals (Xu and van Deventer, 2000; Xu et al., 2001). The leaching results of Na-feldspar in NaOH and KOH solutions (Fig. 3 and 4) show agreement with the observations made on other Al-Si minerals (Xu and van Deventer, 2000; Xu et al., 2001) that NaOH leaches more Al and Si complexes into solution than KOH. Although a significant increase in the concentrations of Al and Si complexes was observed when the concentration of NaOH increased from 2M to 5M, the increase was marginal when the concentration of NaOH increased further to 7.5M. Moreover, it is observed in Fig. 4 that a large increase of leached Al and Si occurred with an increase in the concentration of KOH from 2M to 5M, beyond which the leached Al and Si slightly decreased. The difference between Na-feldspar and the other Al-Si minerals investigated (Xu and van Deventer, 2000; Xu et al., 2001) suggests an effect of inhibition caused the interaction between the alkali metal cations in solution and the Na structured in Na-feldspar (Xu and van Deventer, 2002) during the leaching behavior of Na-feldspar in the 7.5M MOH solutions.

Similar to Na-feldspar, K-feldspar is observed to leach more Al and Si in NaOH than in KOH solution (Fig. 5 and 6). In addition, the leached Al and Si are significantly higher in 5M MOH than in 2M MOH. However, beyond 5M, the increase in the leached Al and Si is inhibited. This inhibitory effect occurs in the leaching of both Na-feldspar and K-feldspar could further affect the geopolymerization of Na-feldspar and K-feldspar in 7.5M MOH solutions.

Effect of alkaline solution on the geopolymerization. The geopolymerization of mixtures of M-feldspar/kaolinite (where M = Na and/or K) can be summarized as the following steps:

- M-feldspar and kaolinite dissolve Al and Si complexes into MOH and silicate solutions to form a gel.
- The formed gel phase becomes thicker and starts polymerizing Al and Si complexes into a three-dimensional amorphous and semicrystalline polymer structure with alkali metal M to compensate the negative charges caused by Al.
- Along with the evaporation of the excess water in the system, the synthesized geopolymer, a mixture of a gel phase and the undissolved M-feldspar and/or kaolinite particles, becomes a monolithic material with a high mechanical strength.

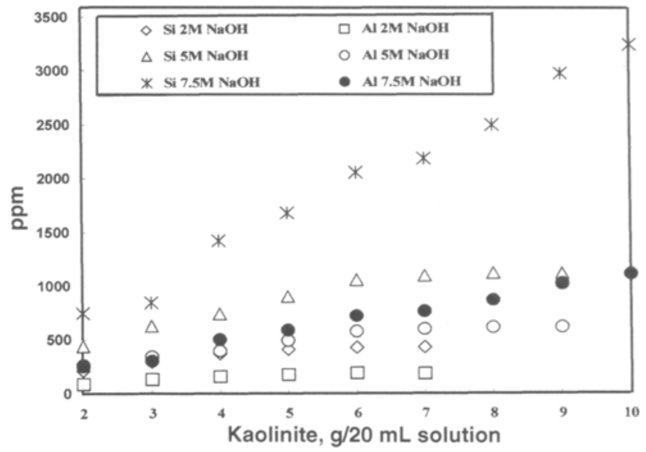

Figure 1 — Leached [Si] (ppm) and [Al] (ppm) of kaolinite in 2M, 5M and 7.5M NaOH solutions.

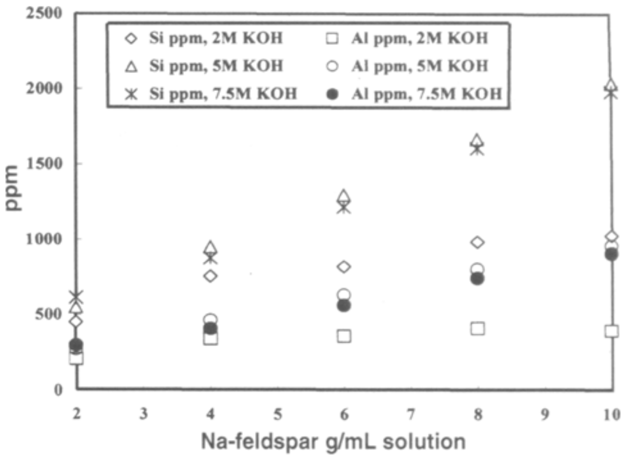

Figure 4 — Leached [Si] (ppm) and [Al] (ppm) of Na-feldspar in 2M, 5M and 7.5M KOH solutions.

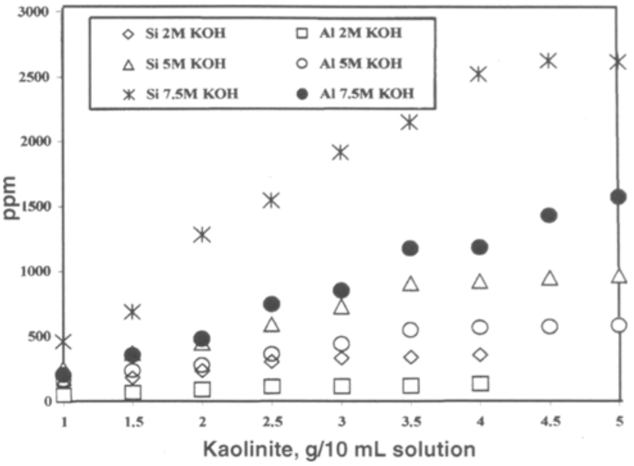

Figure 2 — Leached [Si] (ppm) and [Al] (ppm) of kaolinite in 2M, 5M and 7.5M KOH solutions.

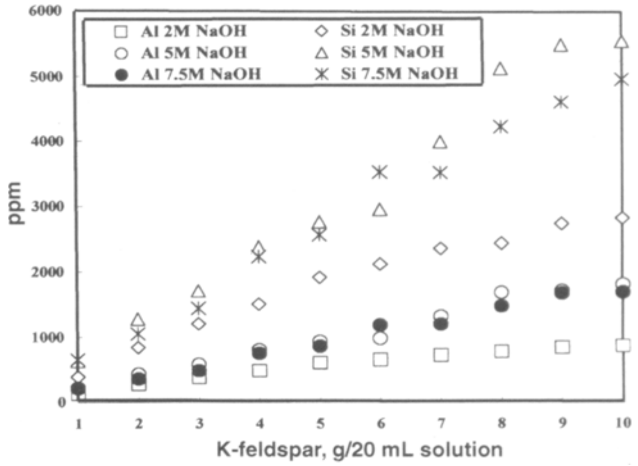

Figure 5 — Leached [Si] (ppm) and [Al] (ppm) of K-feldspar in 2M, 5M and 7.5M NaOH solutions.

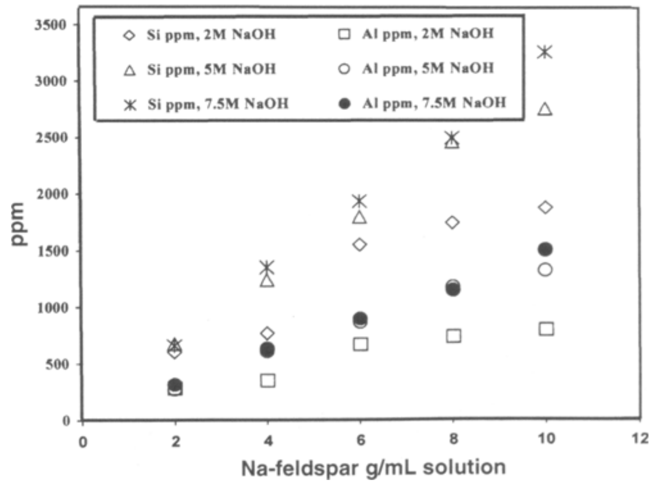

Figure 3 — Leached [Si] (ppm) and [Al] (ppm) of Na-feldspar in 2M, 5M and 7.5M NaOH solutions.

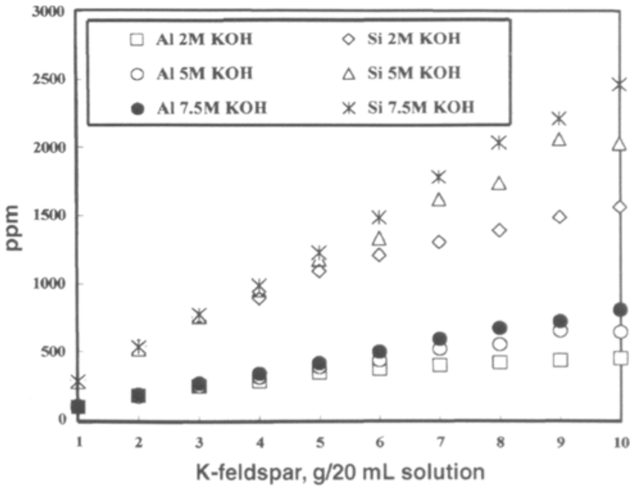

Figure 6 — Leached [Si] (ppm) and [Al] (ppm) of K-feldspar in 2M, 5M and 7.5M KOH solutions.

Therefore, factors that affect any step described above will affect the geopolymerization and consequently the compressive strength of the geopolymers formed.

Figures 7 through 9 depict the compressive strength of geopolymers synthesized from the mixtures of M-feldspar/kaolinite with NaOH and KOH, respectively. The geopolymers formed from K-feldspar/kaolinite with KOH are found to crack to various extents after setting and hardening in an oven at 45°C for 28 days so that no compressive strength data are reported for these samples.

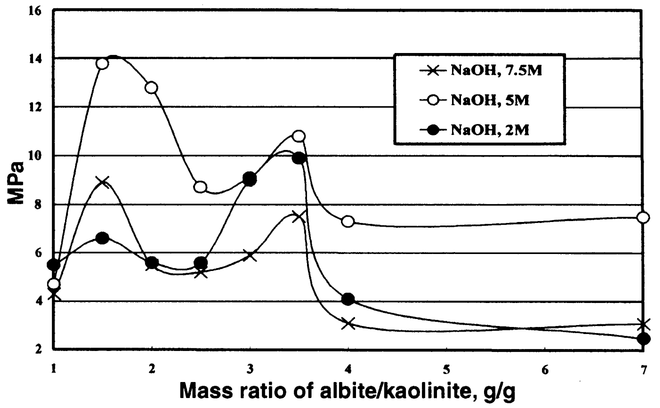

Figure 7 — The compressive strength of geopolymers synthesized from a mixture of Na-feldspar and kaolinite using NaOH.

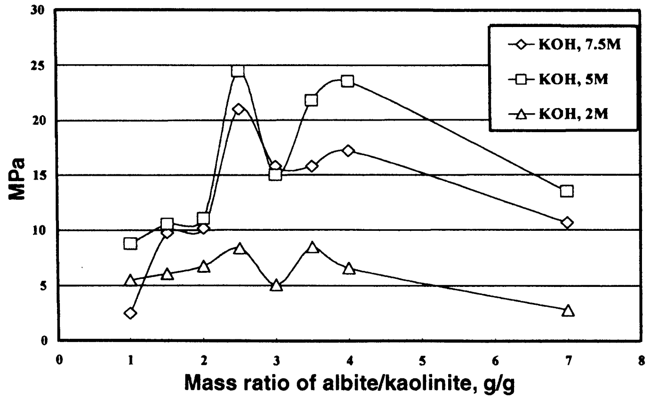

Figure 8 — The compressive strength of geopolymers synthesized from a mixture of Na-feldspar and kaolinite using KOH.

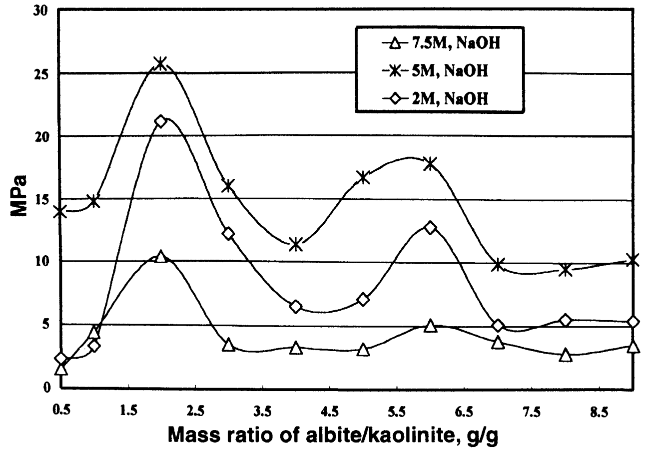

Figure 9 — The compressive strength of geopolymers synthesized from a mixture of K-feldspar and kaolinite using NaOH.

It is noticed from Fig. 7 that geopolymers synthesized at the concentration of 5M show the highest compressive strength followed by the geopolymers synthesized at the concentration of 2M. The concentration of 7.5M produces geopolymers with the lowest compressive strength. The dissolution behavior of Na-feldspar in NaOH solution (Fig. 3) shows that the leached Al and Si are slightly higher in 7.5M solution than in 5M solution. Moreover, both 7.5M and 5M solutions leach substantially more Al and Si than the 2M solution does. Because a higher concentration of leached Al and Si in the solution would result in a geopolymer with a higher compressive strength (Xu and van Deventer, 2000; Xu et al., 2001) the results of Fig. 7 indicate that apart from the dissolution, other factors must dominate the geopolymerization of Na-feldspar in 7.5M NaOH solution.

Table 2 — The viscosity of NaOH and KOH solutions detected at 20°C by a vane rheometer.

NaOH, M	Zero shear viscosity, mPa.s	KOH, M	Zero shear viscosity, mPa.s
2	14	2	11
5	31	5	15
7.5	64	7.5	18

Table 2 lists the viscosity data for NaOH and KOH solutions determined by a vane rheometer. A solution with a higher viscosity means that a longer time period is needed to evaporate the same amount of the excess water from the solution. As noted, the 7.5M NaOH solution possesses a viscosity of 64 mPa.s, which is nearly twice the viscosity of the 5M NaOH solution. This implies that the geopolymerization with 7.5M NaOH solution may require a longer curation period, which can allow the reaction to be fully completed. The study done by Xu and van Deventer (2002) showed that 28 days setting and hardening at 45°C was sufficient for geopolymers synthesized in 5M NaOH solution but 35 days should be applied for geopolymerization taken place in 7.5M NaOH solution. Therefore, it could be the high viscosity of the 7.5M NaOH solution that causes the formed geopolymers (Fig. 7) to soften and have lower compressive strengths than those synthesized in 2M and 5M NaOH solutions.

Figure 8 shows the compressive strength of the geopolymers synthesized from the mixture of Na-feldspar/kaolinite in the KOH solutions. The geopolymers synthesized with 5M KOH possess the highest compressive strength, followed by those synthesized with 7.5M KOH. The geopolymers produced with KOH at the concentration of 2M show the lowest compressive strength. The dissolution of Na-feldspar in the KOH solution, as described in Fig. 4, shows the order of 5M > 7.5M > 2M as well. The correspondence between the trends in the compressive strength of the geopolymers and the extent of dissolution suggests that during the geopolymerization of Na-feldspar/kaolinite with KOH, the leaching extent is the dominant factor which affects the final mechanical strength of the geopolymers.

Figure 9 gives the compressive strength of geopolymers synthesized from K-feldspar/kaolinite with the NaOH solutions. It is found again here that the higher viscosity of the 7.5M NaOH solution significantly affect the geopolymerization with geopolymers synthesized in the 7.5M NaOH solution showing the lowest compressive strength. However, it is also noticed that the highest compressive strength of the geopolymer formed from K-feldspar/kaolinite in the 5M NaOH solution is 25.8 MPa (Fig. 9), which is 86.97% higher than the highest compressive strength obtained from the geopolymer synthesized from Na-feldspar/kaolinite in 5M NaOH solution (Fig. 7). This observation suggests that the molar ratio of Na/K in the gel phase may be another factor that will influence the mechanical properties of the formed geopolymers.

Effects of the mass ratio of M-feldspar/kaolinite on geopolymerization. Besides the general trends of the compressive strength for the geopolymers synthesized from M-feldspar/kaolinite discussed above, two peaks are observed in Figs. 7 through 9. This means that along with an increase in the starting mass ratio of M-feldspar/kaolinite from 1.0, the compressive strength increases until it reaches the first peak. Following a drop, the compressive strength rises again until the second peak and then drops again without any further significant increase occurring. During the geopolymerization of M-feldspar/kaolinite, kaolinite is an Al_2O_3-rich mineral and reacts as an Al_2O_3 supplier so that, along with the increase in the mass ratio of M-feldspar/kaolinite, less kaolinite will be involved in the geopolymerization, consequently reducing the amount of Al_2O_3 in the geopolymeric gel phase and resulting in a gel composition with a higher molar ratio of Si/Al.

An analysis of SEM/EDX on the molar ratio of Si/Al in the gel phases of the geopolymers shows that the molar ratio of Si/Al increases along with an increase in the mass ratio of M-feldspar/kaolinite for all cases studied (Table 3). In the Na-feldspar/kaolinite and K-feldspar/kaolinite systems (Table 3), higher compressive strength is observed for the geopolymers with the Si/Al molar ratio ranging from 2.21 to 2.38 as detected from the Samples 1, 5 and 6 (referring to the first peaks shown in Figs. 7 through 9) in Table 3. The high Si/Al ratio of 4.31 for Sample 8 is associated with an unexpectedly high strength. This demonstrates that the feldspar/kaolinite ratio also plays a role in determining strength. The compressive strength of a geopolymer is believed to be contributed by both the gel phase as a binder and the undissolved particles remaining in the geopolymer as fillers. It is hypothesized that along with an increase in the mass ratio of M-feldspar/kaolinite, the molar ratio of Si/Al contained in the corresponding gel phases will increase to the optimal value or values at which the geopolymers formed show the highest mechanical strength.

Table 3 — The compressive strength and the average molar ratio of Si/Al and Na/K contained in the gel phases of selected geopolymers detected by SEM/EDX analysis (where M = Na and/or K).

Sample No.	M-feldspar/kaolinite, g/g	MOH, M	Compressive strength, MPa	Si/Al, molar ratio
1	Na-feldspar, 1.50	NaOH, 5M	13.8	2.21
2	Na-feldspar, 2.5	NaOH, 5M	8.6	3.35
3	Na-feldspar, 1.0	KOH, 5M	8.8	1.72
4	Na-feldspar, 2.0	KOH, 7.5M	10.3	1.99
5	Na-feldspar, 2.5	KOH, 5M	24.4	2.38
6	K-feldspar, 2.0	NaOH, 5M	25.8	2.26
7	K-feldspar, 4.0	NaOH, 5M	12.5	3.75
8	K-feldspar, 6.0	NaOH, 5M	17.5	4.31

When the mass ratio of M-feldspar/kaolinite is further increased, the molar ratio of Si/Al in the corresponding gel phase increases beyond the optimal value, resulting in a drop in the compressive strength. However, it is observed from Figs. 7 through 9 that the compressive strength of the geopolymers after a drop rises again until it reaches the second peak and then drops afterwards. The presence of the second peak (Figs. 7 through 9) in the compressive strength indicates that some factor other than the molar ratio of Si/Al also affects the compressive strength of the geopolymers. The physical properties given in Table 1 show that the densities of the three minerals, Na-feldspar, K-feldspar and kaolinite are similar, while the hardness of kaolinite is much lower than that for Na-feldspar and K-feldspar. Hence, the change in the mass ratio of M-feldspar/kaolinite will change the average hardness for the solid particles both before and after geopolymerization. Along with an increase in the mass ratio of M-feldspar/kaolinite, fewer kaolinite particles will be involved in the geopolymerization, which increases the probability that kaolinite is transferred completely into the gel phase. The synthesized geopolymers consequently comprise less kaolinite after the geopolymerization. Because the undissolved particles left in a monolithic geopolymer are believed to act as fillers, the fewer kaolinite particles existing in the geopolymers, the higher the average hardness of the undissolved particles will be. Therefore, a formed geopolymer with a higher compressive strength is expected if no or trace kaolinite particles remain undissolved.

Figure 10 presents the XRD patterns recorded for K-feldspar, kaolinite and geopolymers synthesized from K-feldspar/kaolinite at the mass ratios of 5.0 and 6.0, respectively, in the 5M NaOH solution. It is observed from Fig. 10 that the kaolinite characteristic (001) peak is detectable in the geopolymer synthesized at the mass ratio of 5.0. However, this characteristic peak disappeared in the XRD pattern of the geopolymer produced at the mass ratio of 6.0. Such change in XRD patterns implies that at the mass ratio of 6.0, kaolinite particles are completely transferred into the gel phase during the geopolymerization. Referring to Fig. 9, it is noted that at the mass ratio of 6.0, the compressive strength of the corresponding geopolymer reaches the second highest value. Nev-

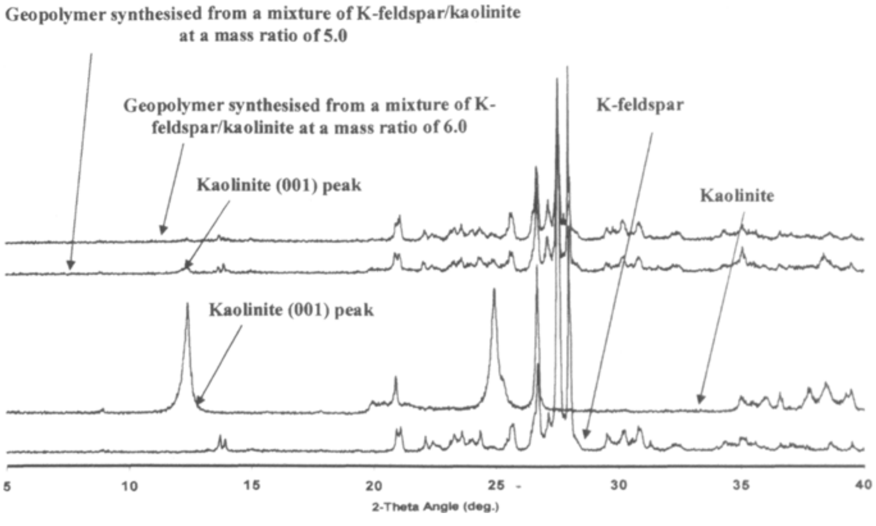

Figure 10 — The XRD pattern of K-feldspar, kaolinite and geopolymers synthesized from a mixture ofK-feldspar/kaolinite at a mass ratio of 5.0 and 6.0, respectively.

ertheless, a further extensive XRD analysis on the geopolymers synthesized from the mixture of K-feldspar/kaolinite in the 5M NaOH solution reveals that the mass ratio of 6.0 is the lowest ratio of K-feldspar/kaolinite for the geopolymers synthesized, with the XRD patterns showing no kaolinite characteristic peaks. Therefore, only the lowest mass ratio of K-feldspar/kaolinite, at which kaolinite is completely transferred into the geopolymeric gel phase, will result in a significant increase in the compressive strength for the geopolymer produced. A series of XRD analyses was conducted separately on the geopolymers synthesized from Na-feldspar/kaolinite. The obtained XRD patterns show that along with an increase in the mass ratio of Na-feldspar/kaolinite, the intensity of the kaolinite characteristic (001) peak becomes lower and lower until completely undetectable. As indicated in the XRD patterns and in Figs. 7 through 9, when geopolymers synthesized at a certain mass ratio show no kaolinite characteristic (001) peak in their XRD patterns, they possess the second highest compressive strengths.

Consequently, it appears that during the geopolymerization of M-feldspar/kaolinite, two factors affect the compressive strength of the geopolymers as the mass ratio of M-feldspar/kaolinite increases. One is the molar ratio of Si/Al contained in the geopolymeric gel phase, and the other is the average hardness of the undissolved particles remaining in the geopolymers.

Cracking has been observed for the geopolymers synthesized from K-feldspar/kaolinite with KOH at all three concentrations. Geopolymerization, as described in Eq. (1), takes place in an alkaline solution with water being an essential reactant. The viscosity of the KOH solution, however, as showed in Table 2, is lower than that of NaOH solution. On the one hand, both K-feldspar and KOH tend to generate a higher geopolymerizing extent, as indicated by leaching in Fig. 5 and 6. On the other hand, a lower viscosity of KOH solution may cause a faster evaporation of water, which is indispensable for geopolymerization. Generally, a fast loss of water from the surface of a sample will quickly hinder an even geopolymerization and will yield a difference in the tension distribution in the sample, eventually causing cracking. Therefore, the quick evaporation of water from the geopolymeric paste could be one of the reasons that the geopolymers synthesized from the K-feldspar/kaolinite in the KOH solution have cracks. This is also the reason that an Al-Si mineral with a higher K_2O content displays poorer geopolymerization properties in KOH than in NaOH as observed previously (Xu and van Deventer, 2000; Xu et al., 2001).

Conclusions

The factors affecting the geopolymerization of Na-feldspar and K-feldspar have been investigated. The results show that both Na-feldspar and K-feldspar have a high dissolution tendency in alkaline solutions (NaOH and KOH) and have a high ability to undergo geopolymerization. Kaolinite, as an Al_2O_3 supplier is observed to enable the geopolymeric gel phase to reach the optimal molar ratio of Si/Al, resulting in a geopolymer with the highest compressive strength. However, excessive kaolinite involved in geopolymerization of M-feldspar/kaolinite negatively affects the final mechanical strength because of its lower hardness. The leaching ability of source minerals (kaolinite, Na-feldspar and K-feldspar) and the viscosity of the alkaline solution are also observed to affect the geopolymerization of M-feldspar/kaolinite. It is expected that a higher concentration of leached Al and Si from source minerals (kaolinite, Na-feldspar and K-feldspar), an appropriate viscosity of the MOH solution with which the source minerals react, an optimal molar ratio of Si/Al in the gel phase and a higher average hardness of the undissolved particles remaining in a geopolymer will generate a geopolymer with the highest compressive strength.

Acknowledgments

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