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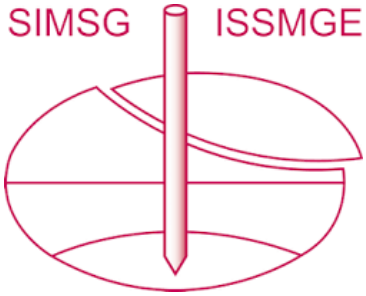

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Utilising novel green binders in ground improvement applications

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ABSTRACT: Special attention is being paid currently to geopolymers as novel binders in ground improvement applications. The use of industrial by-products such as fly ash (FA) and slag (S) in the synthesis of geopolymers makes these alternatives to traditional binders, such as Portland cement, sustainable binders with low-carbon footprint. Geopolymers have been studied and used in a variety of applications, such as concrete or ceramic manufacturing, with controllable conditions of production environment. There are however limited knowledge on the use of geopolymers, as stabilising binders, in ground improvement projects and lack of certainties as to how these new binders would behave in the field where varying factors such as water table or temperature could affect the strength development. This study evaluates the reliability of using a FA and S based geopolymer to stabilise a soft marine clay. The strength development and the mineralogy of the mixtures were studied. The combined FA+S contents were 10, 20 and 30%, and mixtures were prepared at water contents of 0.75, 1.0 and 1.25 liquid limit (LL). Samples were cured for 28 days at temperatures of 10, 25 and 40°C. Strength development was significantly increased by adding the FA+S content, particularly at 20% and higher. Moreover, when the water content was increased from 0.75 to 1.0 LL, strength development was enhanced, followed by a decrease at water content of 1.25 LL. Furthermore, by increasing the curing temperature, higher strengths were achieved and the strength development was accelerated. The results indicated that green geopolymeric binders could be used as reliable binders in ground improvement applications.

1 INTRODUCTION

There is an urgent need to find sustainable alternatives to traditional binders, such as Portland cement, with the increasing growth of population and demand for infrastructure globally. In addition to the consumption of large amounts of natural resources and energy for the production of Portland cement, the emission of CO_2 (around 1 ton CO_2 per ton of Portland cement) and air pollution during the manufacture is notable (Zhang et al., 2013). Attempts have been made, therefore, to find sustainable alternatives, to traditional binders, with a focus on recycling industrial by-products such as fly ash (FA) and slag (S). These attempts have led to the introduction of geopolymers. In geopolymerisation, an alkaline substance is used to activate silica and alumina, abundant in materials such as FA and S, followed by generation of monomers and lastly, polycondensation of these monomers (Cristelo et al., 2013). Geopolymers thus have much lower carbon footprint compared to that of traditional binders as FA and S are abundantly available in the landfills.

In addition to the environmental advantage, better physical and mechanical performance of geopolymers makes them a more attractive alternative com-pared to traditional binders. It has been reported that geopolymers have lower shrinkage, higher compressive and flexural strength, more ductility and more fire and acid attack resistance compared to that of Portland cement (Gao et al., 2013; Zhang et al., 2013; Phoongernkham et al., 2015; Nath & Sarker, 2017).

Considerable amounts of alumina and silica are contained in the structure of clays, such as kaolin, that makes them a suitable source for geopolymerisation; however, the low reactivity of clays due to their layered structure prevents significant strength development through alkaline activation (Heah et al., 2012). The use of calcined materials such as metakaolin, FA and S, to produce geopolymers, was reported to increase the reactivity and achieving considerable strength gain (Xu & Van Deventer, 2002; Hardjito and Rangan, 2005). Relatively high temperatures, more than 600°C, are required albeit to calcine kaolin and turn it into metakaolin (Rovnaník, 2010; Gao et al., 2013), while FA and S, as by-products of electric-

ity generation in power plants and steel manufacturing process, respectively, are already stockpiled in landfills and being produced continuously. Currently, 12.3 tons of FA and 2.6 tons of S are being generated in Australia (ASA, 2016; DEE, 2018), which makes these wastes a sustainable source of geopolymeric binders, at least for the next few decades. In addition, due to having large surface areas, the reactivity of FA and S during alkaline activation is higher compared to metakaolin (Heah et al., 2012).

There has been several studies on the use of geopolymers in applications such as concrete, mortar, brick and ceramic manufacturing (Xu & Van Deventer, 2002; Rovnaník, 2010; Gao et al., 2013; Is-mail et al., 2014; Phoongernkham et al., 2015). The use of geopolymers in ground improvement applications, such as deep soil mixing, on the other hand is quite recent and needs investigation (Cristelo et al., 2013; Zhang et al., 2013; Latifi et al., 2016; Phetchuay et al., 2016; Pourakbar et al., 2016; Singhi et al., 2016; Sukmak et al., 2017; Yaghoubi et al., 2018). Factors such as the binder content, water content and curing temperature have been reported to have significant influence on the strength development of geopolymers. An increase of compressive strength was observed by increasing the content of source of aluminium and silicon, i.e. FA, S and kaolin (Xu & Van Deventer, 2002; Singhi et al., 2016). After a specific curing time at temperatures above 80°C, a decrease in the compressive strength of kaolin, metakaolin and FA+S based geopolymers and water treatment sludge stabilised with FA based geopolymer has been reported previously (Heah et al., 2011; Suksiripattanapong et al., 2015; Park et al., 2016). In a study by Hardjito & Rangan (2005) on FA based geopolymer concrete when heat-cured, no notable further strength development was observed after prolonged curing. In other studies on FA based geopolymer stabilised soft soils, the compressive strengths increased up to one year, even at curing temperature of 85°C (Criado et al., 2007; Cristelo et al., 2011, 2013). The effect of curing temperature on strength development thus is greatly dependent on the type of material. The effect of curing temperatures below the room temperature has been rarely studied (Rovnaník, 2010). Further-more, excessive amounts of water decreased the strength development in geopolymeric concrete and mortar, although the presence of water was essential for geopolymerisation (Hardjito & Rangan, 2005; Gao et al., 2013; Nath & Sarker, 2017).

The manufacturing conditions during the production of concrete, mortar, brick and ceramic are fully or almost fully controllable. In ground improvement projects on the other hand, these conditions can be variable, especially at coastal areas. In Melbourne, Australia For instance, 90% of the time, the ground and air temperatures vary between 10°C to 40°C during the year (Colls et al., 2012; BM, 2016). In addition, different water contents (40-65%), depending on the location and depth, has been reported for a soft marine clay, locally termed as Coode Island silt (CIS). This soft soil covers a wide area and extends to depths of up to 30 m, in Yarra Delta in Melbourne (Ervin, 1992; Phetchuay et al., 2016). Improving engineering properties of this soil is vital since there is a large demand for infrastructures, such as roads and ports, in this area (Phetchuay et al., 2016).

This study aimed at investigating the effect of binder and water content and curing temperature on strength development and changes in the mineralogy of CIS stabilised with FA and S based geopolymers. These were studied through conducting unconfined compressive strength (UCS) and X-ray diffraction (XRD) tests. The results of this study will potentially enable the usage of geopolymers, which consume stockpiled FA and S in landfills, in the ground improvement of soft soils.

2 MATERIALS AND METHODS

2.1 *Materials*

The soft soil, CIS, used in this study was obtained from depths of around 3-5 m in the Port Melbourne region. The natural water content of CIS was in the range of approximately 40-60%. Different properties of CIS, presented in Table 1, were determined by conducting a number of tests in laboratory. The maximum particle diameter (D_{max}) of CIS was 150 µm, and the fine content (particles smaller than 75 µm) was 90%. The liquid limit (LL) and plasticity index (PI) of CIS were 50.4% and 27.0%, respectively. From these results, CIS was classified as a silty clay with high plasticity. The specific gravity (G_s) and pH value of CIS were 2.61 and 7.75, respectively.

Table 1. Properties of the materials

Material	Maximum particle diameter (µm)	Liquid limit (%)	Plasticity index (%)	Specific gravity	pH
Coode Island Silt	150	50.4	27.0	2.61	7.75
Fly ash	106	-	-	2.10	7.50
Slag	63	-	-	2.82	9.50

FA and S were used as the source of alumina and silica, for geopolymerisation. Table 1 presents the characteristics of FA and S. The FA and S had a D_{max} of 106 µm and 63 µm, respectively. The D_{max} of FA and S were specified using a laser particle size analyser, by which the powders were dispersed in air and the particle sizes were detected and measured by laser. This method is useful for measuring the particle size distribution of powders that dissolve or react in

water (Phetchuay et al., 2016). The G_s of FA and S was 2.10 and 2.82, respectively.

X-ray fluorescence (XRF) tests were conducted to determine the chemical composition of CIS, FA and S. From the XRF results, SiO_2 and Al_2O_3 dominated the composition in CIS and FA, whereas in S, CaO was the dominant compound followed by SiO_2 and Al_2O_3.

To evaluate the mineralogy of CIS, FA and S, XRD tests were conducted, and the results are illustrated in Figure 1. The CIS was mainly composed of quartz, illite and feldspar with traces of kaolinite and magnetite. The FA had amorphous phases and was composed of quartz and mullite with traces of hematite and lime. The S was mainly in the amorphous form and traces of gypsum were found in the S.

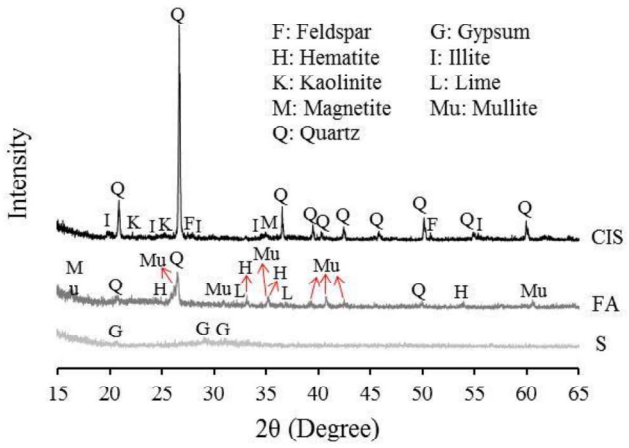

Figure 1. The mineralogy of CIS, FA and S.

A composition of sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3) was prepared as the liquid alkaline activator (L). Initially, NaOH, in the form of beads with 97% purity, was mixed with water to obtain a solution of 8 molarity, and then blended with Na_2SiO_3, in a solution form with a SiO_2/Na_2O ratio of 2, at a NaOH:Na_2SiO_3 ratio of 30:70, as recommended earlier (Cristelo et al., 2011; Phoongernkham et al., 2015; Phetchuay et al., 2016).

2.2 *Methods*

In this study, different combinations of the materials and curing conditions were adopted to replicate and investigate various field conditions. Binder contents of up to 30% are used in typical ground improvement projects (Arulrajah et al., 2009; Horpibulsuk et al., 2011; Pourakbar et al., 2016). In this study hence, combined FA and S contents of 10, 20, and 30% (by dry mass of soil) were used. A mix of FA+S with the FA:S ratio of 25:75 was used in all mixtures as recommended previously. This combination resulted in higher strength development and in early stages of curing, due to the presence of S, as well as achieving an optimum strength development through formation of a coexistence of sodium aluminium silicate hydrate gel and calcium silicate hydrate gel, in the presence of FA. The result of this coexistence is known as calcium sodium aluminium silicate hydrate (CNASH) (Xu & Van Deventer, 2002; Criado et al., 2007; Garcia-Lodeiro et al., 2011; Ismail et al., 2014; Park et al., 2016; Phoongernkham et al., 2015; Phetchuay et al., 2016 Yaghoubi et al., 2018).

For specimen fabrication, initially, the water content of the soil was adjusted to the desired value, to replicate natural water contents in the field. Then, the specific amount of FA+S, mixed in the powder form, was added to the wet CIS. Using a mechanical mixer, the blend was then mixed for 2.5 minutes. After-wards, the specific amount of L (1.0 L/(FA+S) ratio), based on previous findings (Cristelo et al., 2013; Heah et al., 2012; Phetchuay et al., 2016), was added, followed by a further 2.5 minutes of mixing. Specimens with 38 mm diameter and 76 mm height were then prepared by pouring the mixtures in two layers into PVC split moulds. Three specimens were pre-pared for each mixture and curing temperature and the average of the UCS values of three tested specimens was reported as the UCS result.

The testing program is shown in Table 2. The effect of FA+S content, soil water content and curing temperature on strength and mineralogy of mixtures was studied through UCS and XRD testing. FA+S contents of 10, 20 and 30%, and water contents of 0.75, 1.0 and 1.25 LL (of the soil) were considered to prepare the mixtures. Samples were cured at temperatures of 10, 25 (room temperature) and 40 °C for 28 days. Note that 40°C replicates the upper boundary of temperature that is typical in Melbourne region below and above the ground surface (Colls et al., 2012; BM, 2016). Although at deep ground levels, the temperature is not as high as the surface, temperatures of above 32°C have been reported at depths of 0.1-0.5 m in Melbourne (Colls et al., 2012). A 1-mm/min (1.32%/min) rate of displacement was chosen to conduct the UCS tests as specified in AS (2008) and ASTM (2016). After UCS testing, small fractions of the samples, were collected to evaluate the changes in mineralogy of the mixtures. The collected fractions were ground, to have particle sizes smaller than 300 μm, and then air-dried, in an oven set at 50°C, before XRD testing.

Table 2. Testing program.

FA+S content (%)	Water content (LL)	Curing temperature (°C)	Curing time (day)	Test
10, 20, 30	0.75, 1, 1.25	10, 25, 40	28	UCS XRD

3 RESULTS AND DISCUSSIONS

3.1 *Unconfined compressive strength*

The UCS values of the mixtures prepared at different binder and water contents and cured for 28 days are illustrated in Figure 2. The minimum 28-day UCS value (1.034 MPa) recommended for deep mixed cemented columns is used for comparison purposes (Puppala et al., 2008). The minimum UCS requirement was not met in almost all cases when 10% (FA+S) was used and relatively insignificant improvement was obtained. The FA+S in these mixtures was inadequate that led to a poor geopolymeric network through the CIS (Singhi et al., 2016). With increasing the FA+S content to 20 and 30% however, the UCS values increased, possibly due to the in-crease of silicon and calcium in the medium (Xu & Van Deventer, 2002). Monomers were formed by sodium, available in the L together with the dissolved amorphous silicon, aluminium and calcium present in the FA and S. Geopolymeric networks were then formed through the polycondensation of these monomers and resulted in the stabilisation of the CIS, and therefore, the enhancement of UCS (Hardjito & Rangan, 2005; Gao et al., 2013). The silicon and aluminium in the CIS were probably also dissolved and contributed to the strength development, although not significantly due to the low reactivity of soils (Cristelo et al., 2011; Pourakbar et al., 2016).

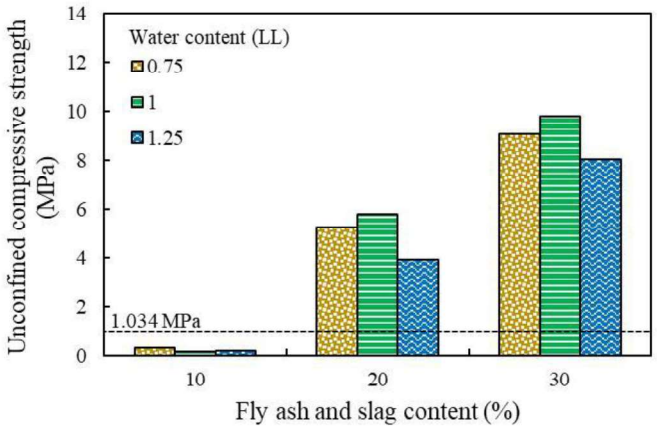

Figure 2. The variation of UCS for soil stabilised with different FA+S and water contents and cured for 28 days.

Increasing the water content decreased the UCS value at 10% binder content (see Figure 2). In these mixtures, the water content was much higher than required and created voids through the structure. In addition, the excessive water reduced the molarity of L, which led to less dissolved aluminium and silicon accessible for geopolymerisation. Thus, inefficient bonding of particles was resulted that caused the creation of a porous structure, with low strengths (Hardjito & Rangan, 2005; Gao et al., 2013; Phetchuay, 2016; Nath & Sarker, 2017). When 20% and 30% (FA+S) were used, at 1.0 LL and 1.25 LL, the UCS values increased and decreased, respectively. The 0.75 LL water content was insufficient for com-plete geopolymerisation, while 1.25 LL was higher than that required, which resulted in lower strengths (Gao et al., 2013). The ideal water content was hence 1.0 LL at 20% and 30% (FA+S) addition. In terms of strength development, the water content variation in the field, within the range of 0.75-1.25 LL, would be safe based on these findings. Overall, CIS + 5% FA + 15% S (20% geopolymeric binder) was found to be the optimum mixture that can be used in high water contents ranging from 0.75 to 1.25 LL.

Figure 3 illustrates the effect of FA+S content and curing temperature on the UCS of the mixtures. At different curing temperatures, the trend of strength development by the variation of binder content was similar to that of changing water contents, explained earlier. The UCS was almost linearly enhanced by increasing the curing temperature from 10°C to 40°C at 20 and 30% (FA+S) content. The curing temperature thus had an accelerating effect on geopolymerisation and strength development (Hardjito & Rangan, 2005; Rovnaník, 2010; Ismail et al., 2014; Suksiripattanapong et al., 2015; Phetchuay et al., 2016). It was reported previously that longer curing times were required at low curing temperatures for geopolymer gels to develop and achieve notable strength developments. The quality of the geopolymers cured at low temperatures were even higher than those cured at higher temperatures, in terms of having less porosity for instance (Rovnaník, 2010; Heah et al., 2011). The results herein indicated that substantial UCS values could be achieved for CIS, containing high water contents, when stabilised with FA and S based geopolymers. These geopolymers were found to be reliable binders to stabilise CIS throughout the year at different temperatures, even at low temperatures during the cold seasons of year, provided relatively long curing times are practicable.

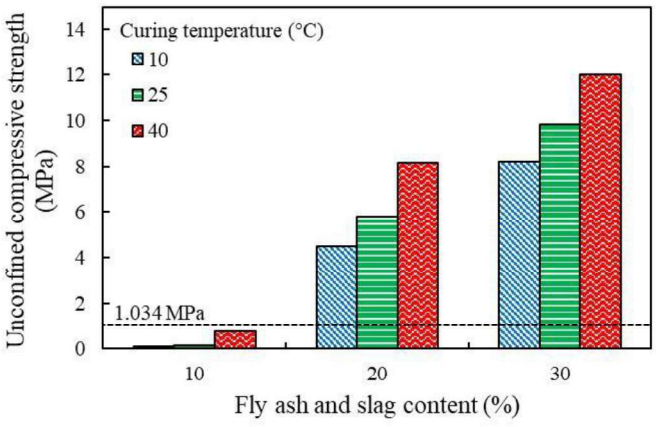

Figure 3. The variation of UCS for soil stabilised with different FA+S contents and cured at various temperatures for 28 days.

3.2 *X-ray diffractometry*

The effect of curing temperature on the crystalline structure of FA and S based geopolymer stabilised CIS was assessed through conducting XRD tests. Fig-ure 4 presents the results of the XRD on CIS stabilised with 20% (FA+S) prepared with initial water content of 1.0 LL and cured at various curing temperatures of 10, 25 and 40°C for 28 days. Except for almost all the quartz and illite at 2θ of 19.7°, the rest of minerals present in the CIS, FA and S were dissolved and amorphous phases were developed (see Figs. 1 and 4). Moreover, the peak intensities of quartz in the mixtures were decreased compared to CIS and FA. This could be attributed to the dissolution of silicon, and subsequently, cementitious reactions, which led to the formation of calcium silicate hydrate as suggested by previous researchers (Criado et al., 2007; Heah et al., 2012; Zhang et al., 2013; Latifi et al., 2016). Figures 4a, b, c show that with increasing the curing temperature, the reduction in the intensities of quartz increased, for instance the quartz at 2θ of 50, which may indicate the contribution of soil particles to the geopolymerisation process (Cristelo et al., 2011; Pourakbar et al., 2016).

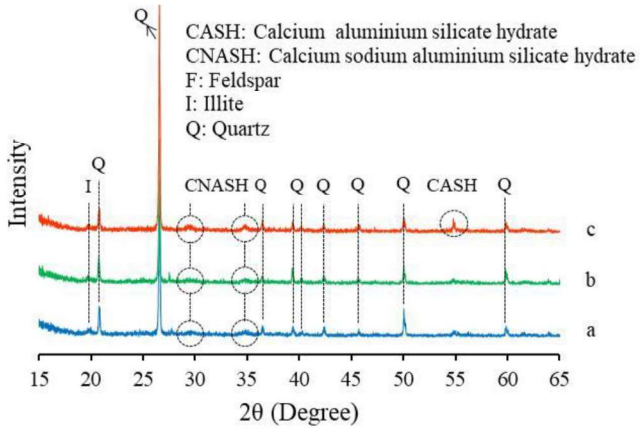

Figure 4. The mineralogy of soil stabilised with 20% (FA+S) prepared at 1.0 LL water content, cured for 28 days at temperatures of: a) 10 °C, b) 25 °C, c) 40 °C.

New phases were developed in the mixtures as a result of these reactions. The new phases at 2θ of around 29-30° and 34-35.5° indicated the formation of calcium sodium aluminium silicate hydrate (CNASH), and at 2θ of 55° indicated the formation of calcium aluminium silicate hydrate (CASH) (Garcia-Lodeiro et al., 2011; Ismail et al., 2014; Suksiripat-tanapong et al., 2015; Latifi et al., 2016; Park et al., 2016; Sukmak et al., 2017). As stated earlier, the ge-opolymerisation process was accelerated with increasing the curing temperature. Park et al. (2016) reported that this could possibly result in the formation of new crystals. The prominent peak formed at 55°, as shown in Figure 4c, which was indexed as a gehlenite hydrate crystal with a CASH nature, might have been resulted from the increase in curing temperature to 40°C (Garcia-Lodeiro et al., 2011; Park et al., 2016). The achieved strength developments was caused by the formation of CNASH and CASH prod-ucts, as a result of geopolymerisation (Phoongernkham et al., 2015; Phetchuay et al., 2016; Sukmak et al., 2017).

4 CONCLUSIONS

The stabilisation of a soft marine clay with novel green geopolymer binders was investigated in this study. Combinations of FA and S in various contents were mixed with the soil and activated by an L. The unconfined compressive strength and mineralogy of the mixtures at various water contents and curing temperatures, which might occur during a typical ground improvement project, were studied. The following results were achieved:

1. Increasing the FA+S content, particularly from 10% to 20%, resulted in significant enhancement of the UCS values due to the geopolymerisation process.
2. Higher UCS values were achieved when the initial water content of the soil was enhanced from 0.75 to 1.0, and increasing the water content further to 1.25 resulted in the decrease of UCS. The reduction in UCS was due to the excessive amount of water that increased the porosity. In addition, by increasing the water content, the molarity of L was reduced that caused the decrease of UCS values.
3. Increasing the curing temperature, from 10°C to 40°C, caused fast precipitation of FA and S and hence accelerated and increased the strength development.
4. From the XRD test results it was found that by increasing the curing temperature, more FA and S were dissolved and CNASH products were formed.

Furthermore, by utilising FA and S based geopolymers in ground improvement projects, where large amounts of binders are required, not only the carbon emission associated with the production of traditional binders would be diminished, but also this could be a solution to the disposal problems of these wastes.

5 ACKNOWLEDGEMENTS

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