OPTIMIZING THE MIX DESIGN OF CLAY BASED GEOPOLYMER CONCRETE

MUHAMMAD MUKHLESUR RAHMAN[1], DAVID W. LAW[2] and INDUBUSHAN PATNAIKUNI[2]

[1] *School of Engineering, RMIT University, Melbourne, Australia and Ministry of Road Transport and Bridges, Bangladesh*
[2] *School of Engineering, RMIT University, Melbourne, Australia*

Although considerable research has been undertaken on alkali-activated binders for geopolymer concrete production, few of these studies have focused on clay based geopolymer concrete. This paper reports the results of an experimental investigation to optimize the mix design of geopolymer concrete prepared solely from clay material. Four Activator Modulus (AM); 1.0, 1.25, 1.5 and 1.75 for each of two Sodium Oxide (Na_2O) dosages of 10% and 15% were selected for this investigation. A total of eight mix ratios comprising 72 specimens were tested. A comparison of physical behavior of clay based geopolymer mortar to that of fly ash based geopolymer mortar has been drawn throughout this paper. A Na_2O dosage of 10% provided consistent strength for all AM tested, however the specimens prepared with Na_2O dosage of 15% showed a reduction with time for all but the AM 1.0 specimens. Compressive strength of specimens prepared with AM of 1 for both of the Na_2O dosage of 10% and 15% exhibited the superiority over other AM. The investigation showed potential for clay material to be an alternative to ordinary Portland cement (OPC).

Key words: Geopolymer, Concrete, Clay, Compressive strength.

1 INTRODUCTION

Geopolymer concrete, an environmental friendly material, has come in to use as a substitute material for OPC concrete in the construction industry in recent times. The production of geopolymer not only limits the emissions of contaminants but also consumes less energy and thus plays a significant role in reducing the global warming problem (Heath *et al.* 2014, Zhang 2013). Different types of raw materials such as fly ash, kaolinite, palm oil fuel ash, rice husk ash, shale, natural zeolite, types of sludge, lignite bottom ash, clay and mud are being used for the production of geopolymer. While discussing the potential of geopolymeric materials Heath *et al.* (2014) pointed out that the use of geopolymer concrete offers a significant advantage in managing the problems associated with aluminosilicate by-products from industrial processes, as well as in reducing the environmental impact arising from the use of OPC concrete. Recent studies reveal that geopolymer concrete made from different industrial waste material display comparable or even better mechanical and durability properties than those associated with OPC concrete (Bernal *et al.* 2012, Ferone *et al.* 2015, Zhang *et al.* 2015).

Research conducted to date indicates that fly ash is the main source of binder material for the production of geopolymer concrete, although other source materials are being investigated. A concern is that fly ash production will decrease in the future as power generation will switch from

coal to other renewable forms of electricity production. Clay, which is a naturally occurring material, and abundant throughout the world, could be a suitable alternate source material to fulfil this requirement.   The potential for global warming could be reduced up to 40% if clay based geopolymer is used as an alternative material of OPC (Heath *et al.* 2014). Perna *et al.* (2014) have also identified the necessity for alternate substitute source material for the production of geopolymer concrete. At present researchers are studying a range of alternate raw materials for geopolymerization such as natural zeolites (Villa *et al.* 2010), calcinated paper sludge (Santa *et al.* 2013), rice husk ash (He *et al.* 2013), lignite bottom ash (Sathonsaowaphak *et al.* 2009). Recently, a new category of geopolymer i.e., blended geopolymer which is derived from the mixture of two or more industrial by-products has drawn attention from a number of researchers (Bhutta *et al.* 2014, He *et al.* 2013, Xu *et al.* 2014). However, research on the utilization of clay as an alternate binder material for geopolymerization system is still in its infancy.

Research carried out in developing clay based geopolymer indicates that this new type of clay based binder possess significant potential to be an alternative material to OPC. This paper reports the results of an experimental investigation to optimize the mix design of geopolymer concrete prepared solely from clay material instead of fly ash. Four Activator Modulus (AM), namely 1, 1.25, 1.5 and 1.75, for each of two Na_2O dosages of 10% and 15% were selected for this investigation. A total of eight mix ratios comprising 72 specimens were tested. AM and Na_2O Dosage is defined in Eq. (1) and Eq. (2) as follows:

$$AM = (SiO_2 \text{ in Alkaline Activator Solution}) / (Na_2O \text{ in Alkaline Activator Solution}) \qquad (1)$$

$$Na_2O \ \text{Dosage (\%)} = (Na_2O \text{ in Alkaline Activator Solution}) / (\text{Mass of Binder}) \qquad (2)$$

2 EXPERIMENTAL PROCEDURE

2.1 Material Used

In this study, the clay material used as a binder material was supplied by The Selkirk Company from the Enfield area about 30 km from Ballarat, Victoria, Australia. The chemical composition along with phase composition and particle size distribution supplied by Selkirk Company are presented in Tables 1 and 2. A mixture of 15 Molar NaOH solution and D-grade liquid Na_2SiO_3 was used as activator solution. River sand of specific gravity of 2.5 and fineness modulus of 3.0 was used as fine aggregate.

Table 1. Chemical composition of clay material.

Oxide	%
Al_2O_3	14.83
SiO_2	69.88
CaO	0.24
Fe_2O_3	3.88
K_2O	2.75
MgO	1.09
MnO	0.015
Na_2O	0.23
TiO_2	0.82
Cl	475 ppm

Table 2. Phase composition of clay material.

PHASE	% Weight
Illite	24.98
Kaolinite	27.17
Quartz	43.05
Rutile	1.67
Chlorite	1.42
Montmorillite	0.17
Albite	0.87
Goethite	0.6

2.2   Proportioning of the Materials

The mass proportioning of ingredients of different AM for both Na_2O dosages of 10% and 15% has been presented in the Tables 3 and 4. The proportioning of ingredients was calculated following an established mix design of Adam (2009). The mass ratio of sand to binder was fixed at 2.75 (ASTM C109/ C109M-07). The w/s (water/solid) ratio of 0.37 was used to ensure consistent workability of the geopolymer mortars. The quantity of water in the mix is the sum of the water contained in the sodium silicate, sodium hydroxide and the added water, while the quantity of solid is the sum of the mass of clay, and the solid contained in the alkaline activator solution.

Table 3. Mix design details based on AM for Na_2O dosage = 15%.

AM	Clay (kg)	Sand (kg)	Na_2SiO_3 (kg)	NaOH (kg)	Added Water (kg)	Na_2SiO_3/NaOH
0.5	1	2.75	0.25	0.36	0.123	0.69
0.75	1	2.75	0.38	0.30	0.096	1.25
1	1	2.75	0.51	0.24	0.07	2.12
1.25	1	2.75	0.64	0.17	0.044	3.61
1.5	1	2.75	0.76	0.11	0.018	6.65
1.75	1	2.75	0.89	0.06	0	14.24

Table 4. Mix design details based on AM for Na2O dosage = 10%.

AM	Clay (kg)	Sand (kg)	Na_2SiO_3 (kg)	NaOH (kg)	Added Water (kg)	Na_2SiO_3/NaOH
0.5	1	2.75	0.17	0.24	0.205	0.70
0.75	1	2.75	0.25	0.2	0.187	1.27
1	1	2.75	0.34	0.15	0.17	2.17
1.25	1	2.75	0.42	0.11	0.153	3.69
1.5	1	2.75	0.51	0.07	0.135	6.95
1.75	1	2.75	0.6	0.04	0.118	14.63

2.3   Specimen Preparation and Testing

The clay was dried in oven at 80^0C for 24 hours. After drying, the clay material was powdered using Ball Mill Grinder Machine. The mill comprised of a total of 15 balls and 5000 cycles were applied to make the clay powder. The powdered clay was sieved through a 106 μm size sieve. This sieved powder was mixed with sand for 4 minutes using 5-liter Hobert mixer. A 15 Molar

NaOH solution and D-grade liquid Na_2SiO_3 of 1.52 g/cc density with composition of 14.7% Na_2O, 29.4% SiO_2 and 55.90% water were premixed 15 minutes before the additional water was added to the activator solution. This activator solution was then added to the mixture of clay powder and sand. After manual mixing for 1 minute, the whole mix was blended by Hobert mixer machine with two rotating speeds: 150 and 300 rev/min for 4 and 2 minutes respectively. The mix was then placed in 50x50x50 mm Teflon moulds followed by 30 second vibration on vibrating table. After allowing 24 hours at room temperature, the moulds were kept in oven at 120^0C for another 24 hours. The moulds were then demoulded and cured at room temperature until testing. Compressive strength measurements of mortars were performed on a TCM Testing Machine in accordance with ASTM C109/C109M-13 with a loading rate of 0.34 $N/mm^2/S$. Three cubes were tested for each data point.

3   RESULTS AND DISCUSSION

The specimens were tested for compression at 7, 14, 28 and 40 days. As a significant fall in strength was observed between 14 and 28 days, the 40 days test was done to assess the rate of strength reduction. Testing at 3 days was not possible as the specimens had not achieved structural integrity. This indicates that clay based geopolymer mortar takes longer duration to harden than that of normal cement mortar or fly ash based geopolymer mortar. A normal Portland cement would be expected to set within 24 hours (Neville 2011), while research using elevated curing for geopolymers has shown that structural integrity is achieved following the elevated curing (Sagoe-Crentsill *et al.* 2010). He *et al.* (2013) also found that red mud (RM) and rice husk ash (RHA) based geopolymer paste took at least 35 days to achieve complete geopolymerization. They mentioned three possibilities: dominant crystalline solid phase acts as unreactive filler, larger particle size slows down the dissolution rate and the presence of impurities may have a detrimental effect on the rate of geopolymerization process. Hanjitsuwan *et al.* (2014) pointed out that at higher NaOH concentration (up to 18 M) the geopolymerization process occurs at a slower rate resulting in a longer setting time.

Table 5. Compressive strength of specimens for Na2O dosage = 15%.

AM	7 days strength (MPa)	14 days strength (MPa)	28 days strength (MPa)	40 days strength (MPa)
1	9.58	10.80	9.40	11.72
1.25	10	11.34	9.74	7.83
1.5	12.86	13.45	9.45	7.64
1.75	20.02	11.96	12.04	11.05

Table 6. Compressive strength of specimens for Na2O dosage = 10%
.

AM	7 days strength (MPa)	14 days strength (MPa)	28 days strength (MPa)	40 days strength (MPa)
1	9.68	14.5	10.42	13.88
1.25	10.87	14.21	10.06	13.25
1.5	11.36	10.97	9.75	12.5
1.75	14.92	10.36	9.96	9.69

The 7, 14, 28 and 40 days compressive strength of geopolymer mortars prepared with Na_2O dosages of 15% and 10% at different AM are presented in Tables 5 and 6. The results show that the strength generally increases from 7 to 14 days for both the 10% and 15% Na_2O dosages. The only exception being the 15%, 1.75 AM which achieves the maximum strength (20.02 MPa) at 7 days which reduces significantly to 14 days (11.96 MPa). The 15%, AM 1.25 and 1.5 both reduced in strength from 14 to 40 days while the AM 1.0 gained approximately a further 1 MPa from 14 to 40 days. Similar behaviour has been observed for FA-rice husk bark ash (RHBA) geopolymer where it was found that the compressive strength of specimens cured at higher temperature(i.e., $> 90^0C$) started to decrease after certain period of time (Nazari *et al.* 2011). They reported that higher temperature destroyed the granular structure of geopolymer which caused dehydration of geopolymer matrix and excessive shrinkage due to contraction of geopolymeric gel. Sukmak *et al.* (2013) observed the existance of micro-cracks at a higher curing temperature (85^0C) with a short curing duration (24 h) and even at a lower curing temperature (75^0C) with a longer curing duration (75 h) for clay-FA geopolymer bricks. Microcracking has also been observed in alkali activated slag geopolymers following elevated curing, which can negatively impact compressive strength (Collins and Sanjayan 2001).

For Na_2O dosage 15%, the 7 days compressive strength was highest (20.02 MPa) for AM of 1.75 whereas AM of 1.5 provided the highest strength (13.45MPa) at 14 days. The compressive strength at 28 days was similar for all the AMs except for AM of 1.75. The 15% Na_2O dosage AM 1.0 provided the highest strength (11.72 MPa) at 40 days. For Na_2O dosage 10%, AM 1.0 showed the optimum strength for 14, 28 and 40 days whereas AM of 1.75 showed the optimum strength at 7 days. A Na_2O dosage of 10% provided more consistent strength than specimens prepared from a Na_2O dosage of 15%. Compressive strength of specimens prepared with AM of 1.0, for both of the Na_2O dosage of 10% and 15%, exhibited superior strength to other specimens. Tables 5 and 6 also show that the compressive strengths at different AM of Na_2O dosage of 10% are greater than corresponding strengths at different AM of Na_2O dosage of 15% except for the AM of 1.75. The results indicate that Na_2O content is having a detrimental effect i.e., the more is the Na_2O content the lower the strength with time. A possible reason may be the formation of Na_2O-CaO-SiO_2-H_2O, as the solubility of Na+ ion is very low in alkali activated binder material as reported by Malolepszy (1993).

4 CONCLUSIONS

The following conclusions were drawn from the above discussion:

- The setting time of clay based geopolymer mortar is longer than that of FA based geopolymer mortars or normal OPC mortars.

- The clay based geopolymers generally showed an increase in strength up to 14 days. After this a slight increase in strength was observed for the specimens with a 10% Na2O dosage, while a decrease was observed for those with a 15% Na2O dosage, other than with an AM of 1.0.

- The results indicate that excess Na2O content has a detrimental effect on the strength gain of clay based geopolymer mortar i.e. the higher the Na2O content, the lower will be the strength with time.

- Na2O dosage of 10% provided consistent strength compared to the specimens prepared from Na2O dosage of 15%.

- Compressive strength of specimens prepared with AM 1.0 for both of the Na2O dosage of 10% and 15% exhibited superiority over other AM.

References

Adam, A. A., Strength and durability properties of alkali activated slag and fly ash-based geopolymer concrete, PhD Thesis, RMIT University, Melbourne, Australia, 2009.

ASTM C109/ C109M-(07-13): Standard Test Method for Compressive Strength of Hydraulic Cement Mortars [Using 50-mm] Cube Specimens.

Bernal, SA., de Gutiérrez, RM., and Provis, JL., Engineering and durability properties of concretes based on alkali-activated granulated blast furnace slag/metakaolin blends, *Construction and Building Materials*, vol. 33, pp. 99-108, 2012.

Bhutta, MAR., Hussin, WM., Azreen, M., and Tahir, MM., Sulphate resistance of geopolymer concrete prepared from blended waste fuel ash, *Journal of Materials in Civil Engineering*, vol. 26, no. 11, p. 04014080, 2014.

Collins, F., and Sanjayan G. J., Microcracking and strength development of alkali activated slag concrete, *Cement and Concrete Composites,* 23(4-5), 345-352, 2001.

Ferone, C., Liguori, B., Capasso, I., Colangelo, F., Cioffi, R., Cappelletto, E.,and Di Maggio, R., Thermally treated clay sediments as geopolymer source material, *Applied Clay Science*, vol. 107, pp. 195-204, 2015.

Hanjitsuwan, S., Hunpratub, S., Thongbai, P., Maensiri, S., Sata, V., and Chindaprasirt, P., Effects of NaOH concentrations on physical and electrical properties of high calcium fly ash geopolymer paste, *Cem Concr Compos,* 45:9–14, 2014.

He, J., Jie, Y., Zhang, J., Yu, Y., and Zhang, G., Synthesis and characterization of red mud and rice husk ash-based geopolymer composites', *Cement and Concrete Composites*, vol. 37, pp. 108-18, 2013.

Heath, A., Paine, K.., and McManus, M., Minimising the global warming potential of clay based geopolymers, *Journal of Cleaner Production*, vol. 78, pp. 75-83, 2014.

Malolepszy J., Some aspects of alkali activated cementitious materials setting and hardening. In *3rd Beijing international symposium on cement and concrete*, vol. 2, 1043–6, 1993.

Nazari, A., Bagheri, A., and Riahi S., Properties of geopolymer with seeded fly ash and rice husk bark ash. *Mater Sci Eng A*, 528(24),7395–401, 2011.

Neville, A. M., *Properties of concrete.* England, Longman Scientific & Technical, 2011.

Perná, I., Hanzlíček, T., and Šupová, M., The identification of geopolymer affinity in specific cases of clay materials, *Applied Clay Science*, vol. 102, pp. 213-9, 2014.

Sagoe-Crentsil, K., Brown T., and Yan S., Medium to Long Term Engineering Properties and Performance of High Strength Geopolymer for Structural Applications, *Advances in Science and Technology*, 9, 135-142, 2010.

Santa, RAAB., Bernardin, AM., Riella, HG., and Kuhnen, NC., Geopolymer synthetized from bottom coal ash and calcined paper sludge, *Journal of Cleaner Production*, vol. 57, pp. 302-7, 2013.

Sathonsaowaphak, A., Chindaprasirt, P., and Pimraksa, K., Workability and strength of lignite bottom ash geopolymer mortar, *Journal of Hazardous Materials*, vol. 168, no. 1, pp. 44-50, 2009.

Sukmak, P., Horpibulsuk, S.,and Shen S-L., Strength development in clay-fly ash geopolymer. *Construction and Building Materials*, 40, 566–74, 2013.

Villa, C., Pecina, E., Torres, R., and Gómez, L., Geopolymer synthesis using alkaline activation of natural zeolite, *Construction and Building Materials*, vol. 24, no. 11, pp. 2084-90, 2010.

Xu, H., Gong, W., Syltebo, L., Izzo, K., Lutze, W., and Pegg, IL., Effect of blast furnace slag grades on fly ash based geopolymer waste forms', *Fuel*, vol. 133, pp. 332-40, 2014.

Zhang, L., Production of bricks from waste materials–a review, *Construction and Building Materials*, vol. 47, pp. 643-55, 2013.

Zhang, Z., Zhu, H., Zhou, C., and Wang, H., Geopolymer from kaolin in China: An overview, *Applied Clay Science,* 2015.