EFFECT OF ELECTROSTATIC REPULSION INDUCED BY SUPERPLASTICIZERS ON THE FLOW BEHAVIOUR OF FLY ASH PASTES

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ABSTRACT

Effect of superplasticizers on fly ash pastes was evaluated by consistency tests (for lower water content pastes) and mini-slump tests (for higher water content pastes). Results indicate that only the polycarboxylate superplasticizers (PCEs) largely improved the flowability of fly ash pastes. Zeta potential measurements were conducted to explain the influence of electrostatic repulsion on flow behaviours of pastes with different superplasticizers. In fly ash pastes, Lignosulfonates (LS) and Polynaphthalene superplasticisers (PNS) depressed the zeta potential to more negative values, while the PCEs increased the zeta potential to less negative values. The more negative zeta potential represents higher electrostatic repulsion between particles. However, the workability results revealed that PCEs had better dispersing ability than PNS and LS. It was deduced that the steric hindrance may be the dominating mechanism for the better dispersing noticed with fly ash pastes, while the electrostatic repulsion may have no impact on dispersing of these pastes.

Keywords: Fly ash paste, superplasticizers, electrostatic repulsion, flowability, zeta potential.

INTRODUCTION

Superplasticizers have been proved to be effective with most of cement pastes by various flow behaviour tests. When added to cement pastes, the superplasticizers get adsorbed on the surface of cement particles whereby the negatively charged ionic sulphonic or carboxylate groups get adsorbed on the positively charged early cementitious compounds (Yang et al., 2019, Colombo et al., 2017). The increased dispersion of particles can be related to the electrostatic repulsion and steric hindrance, which are generated from the adsorbed superplasticizers (Gelardi and Flatt, 2016, Yoshioka et al., 2002, Uchikawa et al., 1997). Electrostatic repulsion results from the negative charge induced by adsorption of superplasticizers (Yoshioka et al., 2002), while steric hindrance results from the thickness of the adsorbed layer (Uchikawa et

al., 1997). A schematic representation of electrostatic repulsion and steric hindrance is presented in Fig. 1.

Even though superplasticizers have been verified to be effective with most of cement types, their behaviour with fly ash is still not clear. Many research projects on zeta potential measurements have been conducted to investigate the working mechanism of superplasticizers in cement pastes. The zeta potential test allows to quantify the electrostatic repulsion caused by adsorbed superplasticizers (Duran et al., 2018). Generally, a high (negative or positive) zeta potential value indicates high electrostatic repulsion between particles (Duran et al., 2018).
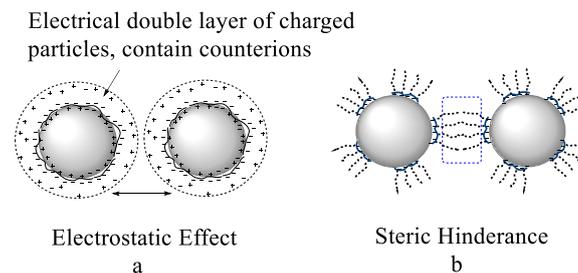

Fig. 1 A schematic representation of electrostatic repulsion (a) and steric hindrance (b), which are generated from the adsorbed superplasticizers on cement surface

The physical characteristics and chemical composition of fly ash are very different from those of cement. Compared to cement, fly ash is spherical in shape, possesses finer grains and lower specific gravity (Bapat, 2012). Unlike cement, the main compounds in Class F fly ash are amorphous glass, quartz, mullite and magnetite. At pH = 7, the zeta potentials of quartz, mullite and magnetite are around -35mV, +10mV and -30mV, respectively (Tang et al., 2002, Erdemoğlu and Sarıkaya, 2006). The presence of both positive and negative ingredients might induce flocculation of fly ash particles.

To more fully understand the interaction between fly ash and superplasticizers, it is necessary to first examine the interaction between fly ash and superplasticizers and compare such interaction with that occurring between cement and superplasticizers. In this regard, the zeta potential measurements could be used to investigate the working mechanism of superplasticizers. Analyses of the changes on zeta potential of particles caused by adding superplasticizers, are of particular importance for defining the scope of fly ash utilisation.

Four different superplasticizers were studied. Two were based on lignosulfonate, polynaphthalene and two were polycarboxylates based. Workability tests were performed to evaluate the dispersing ability of the different superplasticizers. Zeta potential tests on both fly ash and cement were carried out to investigate the change in electrostatic repulsion for different pastes when using different superplasticizers.

MATERIALS AND METHODS

Materials

Fly ash and cement samples, supplied by fly ash Australia Pty Ltd and Cement Australia Pty Ltd, were used in this research. Their chemical composition was determined by X-ray fluorescence (Table 1). The size distribution of fly ash and cement particles were measured using a Malvern Mastersizer 2000 laser diffraction particle size analyser. As shown in Fig. 2, it can be seen that fly ash and cement have quite similar particle size distribution. Compared to fly ash, the distribution of particle sizes of cement is narrower with a smaller median diameter. The crystalline contents in fly ash and cement were identified by X-ray diffraction (XRD) with the Cu Kα radiation at room temperature. The scan range of XRD was 10-70° 2θ values. For the quantitative analysis of chemical compounds in the fly ash and cement, 20% Al_2O_3 was added into the samples as the internal standard. The quantitative analysis results are listed in Table 2.

Table 1. Chemical composition of fly ash and cement

Oxide wt.%	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	CaO	Na_2O	K_2O	SO_3	L.O.I.
Fly ash	60.44	1.05	26.86	3.25	2.56	0.77	0.59	0.09	1.87
Cement	19.56	0.30	4.95	3.04	64.70	0.56	0.22	2.22	3.28

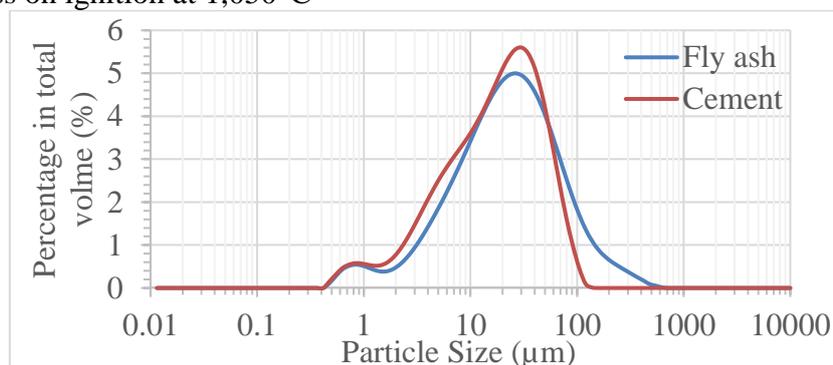L.O.I=loss on ignition at 1,050°C

Fig. 2 Particle size distribution of fly ash and cement[1].

Table 2. The chemical compound content in fly ash and cement

	Fly ash		Cement	
	Content wt. %	SD	Content wt. %	SD
amorphous	74.4	0.8	38.3	2.6
quartz	6.9	0.2		
mullite	18.1	0.4		
magnetite	0.6	0.1		
C_3S			45.8	1
C_2S			0.8	1.1

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[1] The particle size distribution was analysed by dispersing particles in a wet medium (dispersant). The percentage in total volume is the percentage in the total volume of the particles and dispersant.

C$_4$AF		7.2	0.5
C$_3$A		2.7	0.4
bassanite		5.2	1

SD: standard deviation

As shown in Table 1, the fly ash and cement are significantly different in their chemical composition. In fly ash, the SiO$_2$ and Al$_2$O$_3$ are the main oxides. The sum of their percentages is up to 87.3%, which is far more in quantity than other oxides like Fe$_2$O$_3$, CaO, TiO$_2$, etc. In cement, CaO and SiO$_2$ represent the main oxides. The sum of their percentages is up to 84.26%. Due to the high CaO content (64.70 %), cement features high reaction activity in water. Compared to fly ash, cement has a higher crystalline content. C$_3$S, C$_2$S, C$_3$A, C$_4$AF and hemihydrate gypsum CaSO$_4$·0.5H$_2$O (bassanite) represent the main mineralogical phases. As shown in Table 2, the crystalline phases of fly ash and cement are very different. The different composition of crystalline phase in fly ash and cement reflects the different reactivity of the two materials. C$_3$S, C$_2$S, C$_4$AF and C$_3$A in cement could react with water, while quartz, mullite and magnetite in fly ash are non-reactive in water (Singh and Subramaniam., 2017).

The four different superplasticizers used in this research were: a lignosulfonate based superplasticizer (LS), a polynaphthalene based superplasticizer (PNS) and two polycarboxylate based ones (PCE-1 and PCE-2). All the superplasticisers were locally available in the market. Their basic characteristics are presented in Table 3. The solid content, pH value and specific gravity were determined according to ASTM C494. The weight-average molecular weight (M$_w$), number-average molecular weight (M$_n$) and polydispersity index (PDI) were detected by gel permeation chromatography (the detailed procedures are described in Appendix A).

Table 3. Basic characteristics of the different superplasticisers

	Solid content (%)	pH	Specific gravity (g/cm^3)	M$_w$	M$_n$	PDI
LS	39.29	6.03	1.20	57834	44968	1.28611
PNS	41.98	6.59	1.22	159214	139738	1.13938
PCE-1	30.77	5.46	1.06	178543	129705	1.37653
PCE-2	39.27	5.96	1.08	142860	111982	1.27574

Workability Tests
Both normal consistency test and mini-slump test were used to evaluate the workability of fresh pastes. The dosage of superplasticizers were recorded as the mass solid-to-solid ratios of superplasticizer to cement or fly ash.

In the consistency tests, penetration depth of the plunger, in the Vicat Appraratus, was determined according to ASTM C187. The changes in penetration depth caused by using various dosages of different superplasticizers were recorded. Each paste was tested three times, using three separately mixed batches of paste to obtain good statistical reproducibility of the obtained data. A penetration depth–water material ratio curve was drawn for each paste with different superplasticizers at certain dosage.

The normal consistency for each type of mixture was obtained using the respective curve. The normal consistency is the value of water/solid-material ratio for the point that corresponds to the penetration depth of 10 mm. The mass ratio of water was reported to the nearest 0.005.

The mini-slump test was conducted as described by Tan et al (Tan et al., 2017). It was performed using a downscaled Abrams mini-slump cone. The dimensions of the cone are 19 mm (top diameter), 38 mm (bottom diameter), and 57 mm (height). The tests were performed on a flat glass plate. A paper marked with grid squares was put under the flat glass plate. During each test, freshly mixed paste was immediately poured into the cone. After smoothing the top with trowel, the cone was slowly lifted and the paste was allowed to free flow for 1 min. Then, a digital photograph was taken from directly above the paste. The spread area was calculated using the ImageJ software and the spread diameter was then determined by considering the spread area circular. Each paste was tested three times.

Zeta Potential Measurement

The zeta potential measurements were carried out with a Nano-Series Zetasizer (Malvern Instruments Ltd) at 25℃. The zeta potential values of cement particles under the same conditions were also investigated and used as controls. Fly ash and cement pastes were prepared with a solution to solid material ratio of 40:1 with 15 min magnetic stirring. Before each measurement, the suspensions were five times further diluted to adjust the electrical conductivity to lower than 18 mS/cm (Akhlaghi et al., 2017). To obtain good statistical reproducibility of the data, each suspension was tested four times with three runs per measurement.

RESULTS AND DISCUSSION

Consistency Tests

Penetration depth in cement pastes and fly ash pastes with superplasticizers at the dosage of 0.5% are presented in Fig. 3. Curves in Fig. 3 show that cements are better dispersed than fly ash at the same water/solid material ratio. This could be explained in view of the higher reactivity of cement with water. According to Karamanis and Vardoulakis, (Karamanis and Vardoulakis, 2012), the particles will tend to disperse, if they are more strongly attracted to the liquid than to each other. Shift of the penetration depth curve to the left (lower water amount direction) is evidence on whether superplasticizers are working with cement or fly ash. The curves shifted to the direction with lower water cement (fly ash) ratio indicates that the superplasticizer used in paste was more efficient (see in Fig. 3).

In both cement and fly ash pastes, the dispersing abilities of superplasticizers are as follows: PCE-2>PCE-1>PNS>LS. Even though the LS was less effective with the cement pastes at the dosage of 0.5%, it still improved the workability of cement pastes. However, in fly ash pastes, LS was the only superplasticizer that led to poor workability compared to the control paste that did not have any superplasticiser.

In order to better check the influence of different superplasticizers at different dosages, the normal consistency was determined from different penetration depth curves like those shown in Fig. 3. The results are listed in Table 4. It is shown in Table 4 that for the cement pastes, both of the superplasticizers main types exhibited a water reducing capability in all dosages, except for the LS with dosage of 0.1% whose water reducing capability decreased by 4.94%. This may be because the dosage of 0.1% is much lower than the optimal dosage of LSs. The normal consistency of cement paste decreased with the increasing dosage of superplasticizers. However, this tendency was not similar with examining the effect of superplasticizers on normal consistency of fly ash pastes. It is clear that LS increased the value of normal consistency of fly ash paste while PNS only slightly reduced the normal consistency of fly ash paste. In contrast, the decreasing in the normal consistency of fly ash caused by PCE-1 and PCE-2 were very clear.
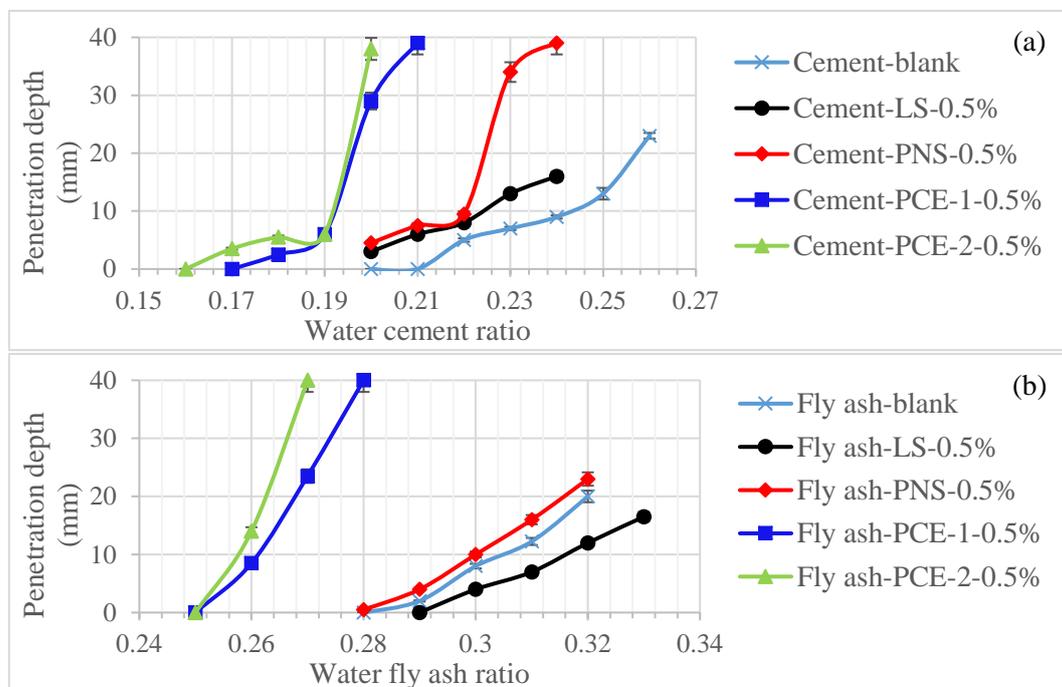

Fig. 3 Penetration depth versus water binder ratio in cement pastes (a) and fly ash pastes (b) with different superplasticizers at the dosage of 0.5%.

Table 4. Effect of different superplasticizers on the normal consistency of cement pastes and fly ash pastes

	cement				fly ash			
Dosage	0.00%	0.10%	0.30%	0.50%	0.00%	0.10%	0.30%	0.50%
LS		0.255	0.225	0.225		0.310	0.305	0.315
PNS	0.245	0.235	0.230	0.220	0.305	0.300	0.300	0.300
PCE-1		0.240	0.210	0.190		0.280	0.270	0.260
PCE-2		0.225	0.210	0.190		0.275	0.260	0.260

Mini-Slump Test

Different water/solid-material ratio were used to obtain the blank mix with the spread diameter of 50 ± 2 mm and this was the starting point achieved by all blank (no superplasticiser) pastes. In the test, the water/solid-material ratio were 0.4 for fly ash pastes and 0.37 for cement pastes.
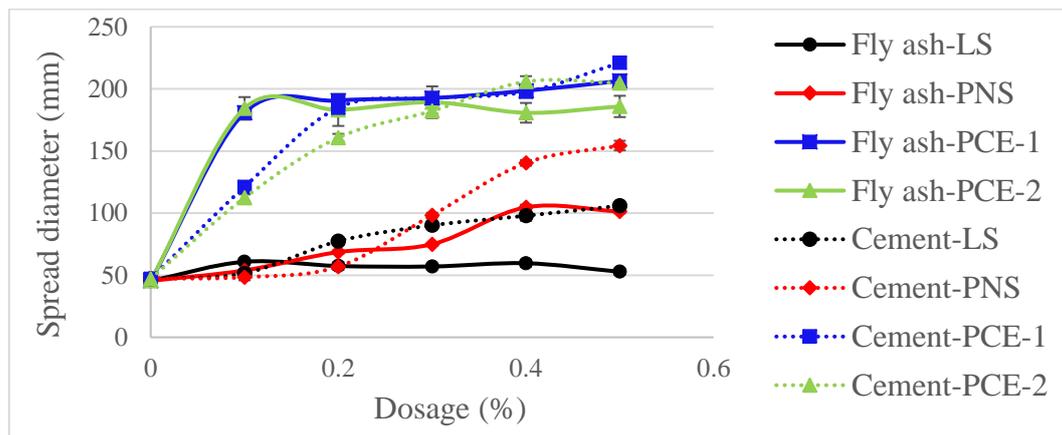

Fig. 4 Variation in spread diameter enhancement by using different dosage of superplasticizers.

Fig. 4 reports the spread diameters of both fly ash and cement pastes with different dosage of superplasticizers by the mini-slump test. It can be seen that the PCEs could largely improve the spread diameter of both cement pastes and fly ash pastes. For fly ash pastes, both the PCE-1 and PCE-2 induced a significant increase of the diameter passing to 133.3 mm and 140.7 mm respectively, at the dosage of 0.1%. On the contrary, cement pastes containing PCE-1 and PCE-2 at the same dosage end up in increments of only 73.5 mm and 64.3mm, respectively. This may have been caused by the intercalation (trapping) of long side chains of PCEs into the hydration products with layered structure during the early hydration stage (Plank et al., 2006). Due to the consumption by intercalation, the amounts of effective PCEs in cement pastes were reduced (Plank et al., 2006). Therefore, the increments in spread diameter of cement pastes caused by adding superplasticizers were limited at low dosage. This limitation may be eliminated at higher dosage. When the dosage of PCEs exceeds 0.3%, the increments in spread diameter of cement pastes were larger than that of fly ash pastes. Compared to PCEs, the increments induced by adding PNS and LS were much smaller. Even though the PNS and LS were less effective than PCEs, the spread diameter increased with the increasing dosage of PNS and LS from 0.1% to 0.5%. However, the tendency for the fly ash pastes was not the same. A saturation point appeared at the dosage of 0.4% for PNS and the spread diameter slightly decreased with the increasing dosage of LS.

Zeta Potential Measurement

The electrostatic repulsion between particles could be determined by measurements of the zeta potential. Higher average absolute zeta potential value represents larger

electrostatic repulsion force between particles. The zeta potential was measured for each superplasticizer at increasing dosages from zero to 11mg/g of solid binder. The measured results are shown in Fig. 5. The changing of the zeta potential to more negative values indicates increasing electrostatic repulsion between particles (Plank and Sachsenhauser, 2006). If the zeta potential changes slightly during the addition of superplasticizers, steric hindrance would be the dominating mechanism for dispersion (Plank and Sachsenhauser, 2006).
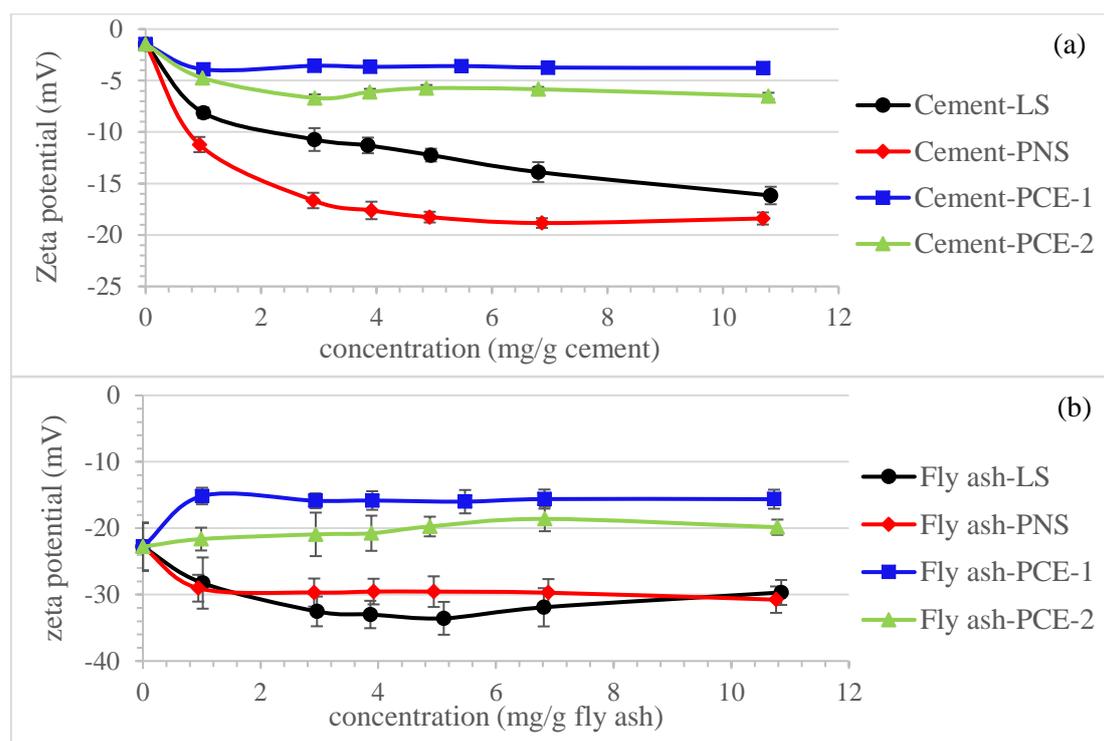

Fig. 5 Zeta potentials of cement (a) and fly ash particles (b) in aqueous solution containing different superplasticizers as a function of superplasticizer dosage

As it is presented in Fig. 5a, in the case of cement, all superplasticizers decreased the zeta potential to more negative values. Therefore, the effect of electrostatic repulsion in the dispersion model was confirmed. The electrostatic repulsion effect was as follows: PNS > LS >> PCE-2 > PCE-1. However, the results in workability test revealed that the dispersion effect followed: PCE-1 > PCE-2 > PNS > LS (see Fig. 4). These results indicated that the electrostatic repulsion can not be the only mechanism for the dispersing ability of superplasticizers. The dispersing ability of PCEs was higher than PNS and LS, but the increase in negativity of zeta potential caused by adding PCEs was lower than those measured using PNS and LS. The slight change in zeta potential revealed that steric hindrance may be the dominating mechanism for PCEs to disperse cement (Plank and Sachsenhauser, 2006). Both types of superplasticizers are negatively charged polymers, thus the adsorption of superplasticizers would lead to a more negative zeta potential. The limited change of zeta potential caused by adding PCEs indicated that the increased negative charge was

'sheltered'. In relation to the structures of different superplasticizers, the 'shelter' of charges would result from the shift of the shear plane[2] to a location far away from the particle surface. Due to the adsorption of PCEs, the shift of the shear plane may be caused by the larger adsorbed layer thickness. Generally, the potential in solution is decreased with the distance away from the charged particles. The side chains of PCEs are non-charged, therefore the potential would decrease with the distance away from the particles. As a result, the changes of zeta potential caused by adding PCEs were limited. According to the changes of zeta potential, it was suspected that the side chains of PCE-1 are longer than those of PCE-2. The longer side chains would produce higher steric hindrance and also lead to less zeta potential change for the particles. As it is shown in Fig. 1b, the steric hindrance results from the thickness of the adsorbed layer. The limited change in zeta potential indicated that the electrostatic repulsion provided by PCEs was very low. However, the the thickness of the non-charged adsorbed layer is considerable. The presence of steric hindrance could be the explanation for the less increase in negativity of particles with added superplasicizer. The higher workability, therefore has resulted from the steric hindrance effect rather than the electrostatic repulsion dominant in the PNS and LS cases. For the PNS and LS, the significant increase in negativity, results in higher workability. Both of the PNS and LS lack side chains. Thus, it can be concluded that the electrostatic repulsion may be the dominating mechanism of cement dispersion for PNS and LS. These results accord with the theory proposed by other researchers (Łaźniewska-Piekarczyk, B., 2012).

The effect of superplasticizers on the zeta potential of fly ash particles is presented in Fig. 5b. When superplasticizers were added into the pastes, LS and PNS depressed the zeta potential to more negative values, but the PCE-1 and PCE-2 increased the zeta potential to less negative values. However, the dispersing ability of LS was the worst according to the result of workability, followed by PNS. In contrast, PCE-1 and PCE-2, which caused a change of the zeta potential towards more positive values, significantly improved the flow ability of fly ash pastes even at the dosage as low as 0.1%. Due to the less negative zeta potential, the electrostatic repulsion that is needed for dispersing fly ash particles plays a very limited role in the dispersing mechanism of PCEs. The steric hindrance is instead, the most likely agent for this mechanism. Thus, when mixing with superplasticizers, the steric hindrance may be the dominating mechanism for the better dispersing of fly ash pastes, while the electrostatic repulsion may have no impact on dispersing fly ash pastes. The details about how the steric hindrance affects the dispersing behaviour will be proposed in the next section.

Adsorption Model
The adsorption formations of superplasticizers on fly ash particles are shown in Fig. 6. Due to the highly negative charge density of LS and PNS, it is possible for LS and PNS to get adsorbed on the positive part of fly ash and make the zeta potential more negative. However, turning the fly ash to more negatively charged particles have no impact on increasing the flowability of fly ash pastes (see in Fig. 3b and Fig. 4). In

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[2] When a colloid particle moving under electric field, a shear plane always develops near the surface of a moving particle. The potential at the shear plane is the zeta potential (ζ) we can deteremined.

most cases, the increase in negativity of zeta potential of fly ash particles results in lower workability. The explanation for this phenomenon may be that the adsorption of one polymer molecule with multi fly ash particles is due to the highly negatively charged density of LS and PNS, as is presented in Fig. 6a. The flocculation appears when multi particles are adsorbed by only one molecule. The combination of serval particles can also decrease the zeta potentials to a more negative value. For the PCE-1 and PCE-2, only a decrease in absolute zeta potential value was detected (see in Fig. 5b). This is different to their behaviour in cement pastes. The explanation for the difference of the change in different pastes is the formation of different adsorption formation on the different particles. As presented in Fig. 6b, the PCE-1 and PCE-2 may get perpendicularly adsorbed on the fly ash particles. Due to the low positive charge density and scattered positive charge distribution of fly ash particles, flexible PCEs cannot easily get adsorbed with a "flat" formation as shown in Fig. 1. With the perpendicular adsorption, sufficient anionic exposures on the surface of the polymer layer, result in further adsorption of cations on the polymer layer surface. In this way, particles' zeta potential will become less negative.
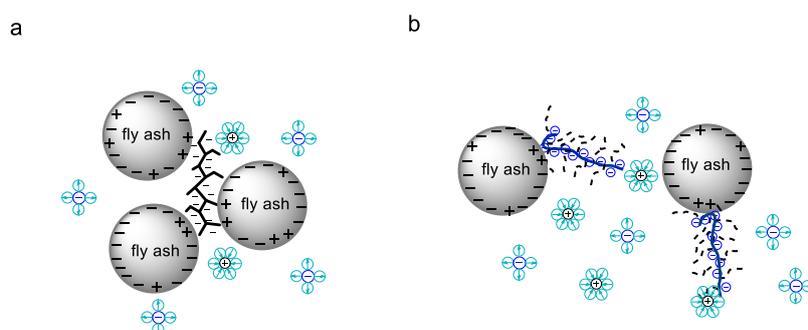

Fig. 6 Illustration of the adsorption conformation on fly ash particles for LS and PNS (a) and PCEs (b).

CONCLUSION

The dispersing ability of superplasticizers in fly ash and cement pastes was evaluated by workability tests. It has been found that polycarboxylate based superplasticizers were more efficient than lignosulfonate and polynaphthalene based superplasticizers for both types of pastes. In fly ash pastes, the dispersing ability of lignosulfonate and polynaphthalene based superplasticizers was limited, especially for the pastes with lower water to fly ash ratios.

Zeta potential measurements were carried out to gain more insight into the interaction between the superplasticizers and the fly ash particles. When superplasticizers were added into the pastes, LS and PNS depressed the zeta potential to more negative values, but the PCEs increased the zeta potential to less negative values. The results indicated that the LS and PNS would induce significant electrostatic repulsion, while the electrostatic repulsion provided by PCEs was very low.

Zeta potential measurements were useful also for understanding the nature of superplasticiser function in both fly ash and cement pastes and to relate the values of that potential to the types of superplasticizers and adsorption formations of the superplasticisers in those pastes.

So far, the results indicated that the dispersing ability of superplasticizers in fly ash paste is greatly dependant on the steric hindrance. Adsorbed formation also have great influence on the dispersing ability of superplasticizers. Further measurements like adsorption amount and thickness of adsorbed layer will be conducted to further investigate the behaviour between superplasticizers and fly ash particles.

APPENDIX A: Gel Permeation Chromatography Analysis (GPC)

The average molecular weight and distribution of the SPs were determined by GPC using a shidmazu GPC unit running on Cirrus GPC software. The flowrate is 0.8mL/min and the eluent is water with 0.02% sodium azide solution. The equipment is calibrated with Polyethylene glycol standard molecular weight. With this meansurement, the weight-average molecular weight (M_w), number-average molecular weight (M_n) and polydispersity index (PDI) would be obtained.

M_n is just the total weight of all the polymer molecules in a sample, divided by the total number of polymer molecules in a sample. M_w is based on the fact that a bigger molecule contains more of the total mass of the polymer sample than the smaller molecules do.

$$M_n = \frac{\sum M_i \cdot n_i}{\sum n_i} \qquad \text{Eq. 1}$$

$$M_w = \frac{\sum M_i \cdot w_i}{\sum w_i} = \frac{\sum M_i^2 \cdot n_i}{\sum M_i \cdot n_i} \qquad \text{Eq. 2}$$

In which, M_i is the molecular weight of a chain and n_i is the number of chains of that molecular weight.

PDI is used to evaluate the broadness of a molecular weight distribution of a polymer. The PDI is defined as,

$$\text{PDI} = \frac{M_w}{M_n} \qquad \text{Eq. 3}$$

The larger the polydispersity index, the broader the molecular weight. For the monodisperse polymers, the PDI is equal to 1.

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REFERENCES

Akhlaghi, O., Aytas, T., Tatli, B., Sezer, D., Hodaei, A., Favier, A., Scrivener, K., Menceloglu, Y. Z., and Akbulut, O. (2017). "Modified poly(carboxylate ether)-based superplasticizer for enhanced flowability of calcined clay-limestone-gypsum blended Portland cement." *Cement and Concrete Research*, 101, 114-122.

Bapat, J. D. (2012) *Mineral admixtures in cement and concrete*, CRC Press, Boca Raton.

Colombo, A., Geiker, M., Justnes, H., Lauten, R. A., and De Weerdt, K. (2017). "On the mechanisms of consumption of calcium lignosulfonate by cement paste." *Cement and Concrete Research*, 98, 1-9.

Duran, A., González-Sánchez, J., Fernández, J., Sirera, R., Navarro-Blasco, Í., and Alvarez, J. (2018). "Influence of Two Polymer-Based Superplasticizers (Poly-naphthalene Sulfonate, PNS, and Lignosulfonate, LS) on Compressive and Flexural Strength, Freeze-Thaw, and Sulphate Attack Resistance of Lime-Metakaolin Grouts." *Polymers*, 10(8), 824.

Erdemoğlu, M., and Sarıkaya, M. (2006). "Effects of heavy metals and oxalate on the zeta potential of magnetite." *Journal of Colloid and Interface Science,* 300(2), 795-804.

Gelardi, G. and Flatt, R.J. (2016). *Science and Technology of Concrete Admixtures*, Woodhead Publishing, Cambridge.

Karamanis, D., and Vardoulakis, E. (2012). "Application of zeolitic materials prepared from fly ash to water vapor adsorption for solar cooling." *Applied energy*, 97, 334-339.

Łaźniewska-Piekarczyk, B. (2012). "The influence of selected new generation admixtures on the workability, air-voids parameters and frost-resistance of self compacting concrete." *Construction and Building Materials*, 31, 310-319.

Plank, J., Dai, Z., Zouaoui, N., and Vlad, D. (2006). "Intercalation of polycarboxylate superplasticizers into C_3A hydrate phases." *Special Publication,* 239, 201-214.

Plank, J., and Sachsenhauser, B. (2006). "Impact of molecular structure on zeta potential and adsorbed conformation of α-allyl-ω-methoxypolyethylene glycol-maleic anhydride superplasticizers." *Journal of advanced concrete technology,* 4(2), 233-239.

Singh, G. B., and Subramaniam, K. V. (2017). "Evaluation of sodium content and sodium hydroxide molarity on compressive strength of alkali activated low-calcium fly ash." *Cement and Concrete Composites*, 81, 122-132.

Tan, Z., Bernal, S. A., and Provis, J. L. (2017). "Reproducible mini-slump test procedure for measuring the yield stress of cementitious pastes." *Materials and Structures,* 50(6), 235.

Tang, Y. F., Lu, Y. N., Li, A. D., Ling, H. Q., Wang, Y. J., and Shao, Q. Y. (2002). "Preparation of composite coating particles of α-Al_2O_3–SiO_2 by heterogeneous nucleation-and-growth processing." *Materials Letters,* 56(4), 446-449.

Uchikawa, H., Hanehara, S., and Sawaki, D. (1997). "The role of steric repulsive force in the dispersion of cement particles in fresh paste prepared with organic admixture." *Cement and Concrete Research,* 27(1), 37-50.

Yang, Z., Yu, M., Liu, Y., Chen, X., and Zhao, Y. (2019). "Synthesis and performance of an environmentally friendly polycarboxylate superplasticizer based on modified poly (aspartic acid)." *Construction and Building Materials*, 202, 154-161.

Yoshioka, K., Tazawa, E. I., Kawai, K., and Enohata, T. (2002). "Adsorption characteristics of superplasticizers on cement component minerals." *Cement and Concrete Research,* 32(10), 1507-1513.