ADDITION OF FILLERS IN CEMENTITIOUS PASTE FOR IMPROVING PACKING DENSITY AND WATER FILM THICKNESS

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ABSTRACT. The packing density of the cementitious paste phase has great influence on the fresh and hardened properties of the mortar and concrete. At any water/binder (W/B) ratio, a higher packing density could increase the amount of excess water for lubricating the cementitious paste and improve the rheology of the mortar and concrete. For any given rheological and workability requirement, the W/B ratio can be reduced to improve the strength of the mortar and concrete. The improvement of packing density can also result in more excess water to coat the solid particles and increase the water film thickness (WFT). This research aims to investigate the effects of addition of fillers on the packing density and WFT of the cementitious paste. The WFT was evaluated based on the measured data of packing density and specific surface area. The experimental results proved that the addition of fillers and superplasticizer can significantly improve the packing density and WFT for enhancing the mortar and concrete performance.

Keywords: Cementitious paste, Filler, Packing density, Superplasticizer, Water film thickness

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INTRODUCTION

The cementitious paste phase of mortar and concrete generally consists of cement, cementitious fillers and inert fillers. The packing density of the paste has great influence on the fresh and hardened properties of the mortar or concrete mix [1,2]. This is because the water in the paste has to first fill up the voids among the solid particles in the paste, and then the excess water (water in excess of that needed to fill the voids) lubricates the paste and imparts flowability to the mortar or concrete mix. Therefore, at any fixed water/binder (W/B) ratio for achieving given strength requirement, a higher packing density of binder would release more excess water to improve the rheology [2-4]. Experimental investigations have revealed that the flowability of cement paste increased with the packing density of the solid particles [5,6]. For any given rheological and workability requirement, the strategy of increasing packing density may be adopted to improve the mortar or concrete strength through adopting a lower W/B ratio. Such approach has been employed to produce very high-strength mortar and concrete in the 1990s [7,8]. Hence, improving the packing density of cementitious paste is pivotal to the production of high-performance mortar and concrete.

In assessing the effect of improvement in packing density, the concept of water film thickness (WFT) can be used [9]. Basically, the WFT is the average thickness of water film coating the solid particles in the paste, and is evaluated as the amount of excess water divided by the surface area of the solid particles. The packing density and WFT are highly dependent on the particle size distribution of the solid in paste phase. If the particles are well graded, the packing density can be increased by successive filling of voids by smaller size particles. In other words, the medium size particles would fill the voids between the larger size particles, whereas the smaller size particles would fill the voids between the medium size particles, so that the overall volume of voids could be reduced. For this reason, the addition of micro-fine fillers whose size is finer than cement can possibly improve the packing density. Another key to desirable particle packing is to avoid the agglomeration of fine particles. Superplasticizer (SP) can be added to the fresh paste during mixing for this purpose. It should be noted that the wet packing test method needs to be employed to account for the effects of SP, which is a dispersant functioning in the water-solid mixture.

The current research aims at investigating the effects of addition of cementitious and inert fillers on the packing density and WFT of the cementitious paste. An experimental programme has been launched to study the packing density of cementitious pastes with various SP dosages and blended with different types of fillers including superfine cement (SFC), condensed silica fume (CSF) and limestone powder (LSP). For each mix, the wet packing density was measured, and the WFT was evaluated based on the measured data of packing density and specific surface area. Through the experimental investigation, the positive effects of adding fillers and SP in improving the packing density and WFT have been revealed.

EXPERIMENTAL METHOD

Regarding the materials, the cement used was ordinary Portland cement (OPC) conforming to European Standard EN 197. Both cementitious and inert fillers are encompassed in the experimental programme. For cementitious filler, superfine cement (SFC) supplied from France and condensed silica fume (CSF) supplied from Norway were used. For inert filler, limestone powder (LSP) supplied from China was used. The solid densities of the OPC, SFC, CSF and LSP were measured in accordance with British Standard BS 4550: Part 3 and their particle size distributions were measured using a laser diffraction particle size analyser. Table 1 summarises the results of solid density and volumetric mean particle size and Fig. 1 depicts the particle size distributions of the materials. It is noteworthy that all the fillers used were finer than the cement. The volumetric mean particle size of the SFC and CSF was respectively finer than 1/4 and 1/50 of that of the OPC, whereas the LSP was marginally finer than the OPC. A polycarboxylate-based SP in aqueous state conforming to European Standard EN 934 was employed. The molecular structure of the SP contained a backbone of active-monomer and multiple side chains of graft copolymers [10]. It can disperse fine particles by both electrostatic repulsion and steric hindrance. The solid mass content of the SP was 20% and the relative density was 1.03.

Table 1 Solid density and volumetric mean particle size of materials

MATERIAL	OPC	SFC	CSF	LSP
Solid density (kg/m^3)	3112	2940	2196	2642
Volumetric mean particle size (μm)	12.5	3.1	0.2	10.3

Regarding the mix design, the SP dosage was varied among 0%, 0.5%, 1%, 2% and 3% by mass of OPC in order to determine the optimum SP dosage, which is taken as the saturation dosage in this study. In subsequence, with the adoption of saturation SP dosage, the SFC content was varied from 0% to 30% in increments of 10% by mass of (OPC+SFC), the CSF content was varied from 0% to 25% in increments of 5% by volume of (OPC+CSF), the LSP content was varied from 0% to 20% in increments of 5% by mass of (OPC+LSP). The CSF content was designated on volumetric basis in view of the large difference of densities among OPC and CSF, and it is the solid volume rather than the mass that influences the particle packing. The W/B ratio by mass of the above blended paste mixes was varied to investigate the respective WFT.

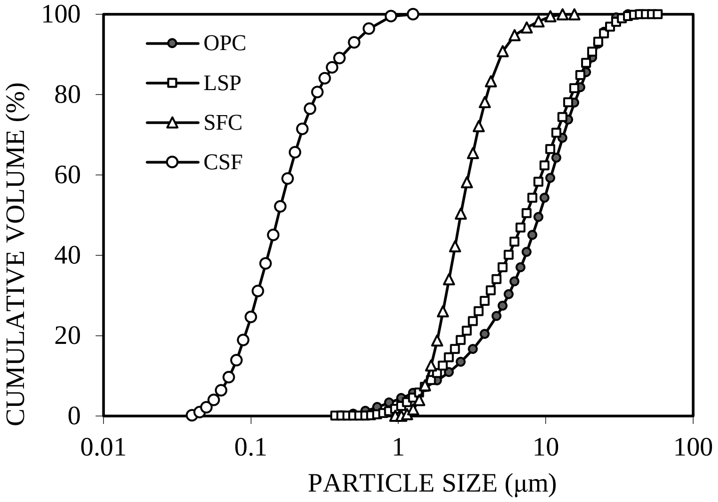

Figure 1 Particle size distributions of materials

The packing density was measured using the wet packing test method as described in detail in reference [9]. In contrast with the dry packing method, the wet packing method can cater for the effects of SP by adding the same amount of SP during the sample preparation. To carry out the measurement, the binder materials at predetermined proportions were mixed with water at different W/B ratios, and the solid concentration ϕ of each paste mix was determined. The W/B ratios ranged from insufficient to more than sufficient water to fill the void space between solid particles. It can be observed that at relatively low W/B ratios, ϕ increased with the W/B ratio, while at relatively high W/B ratios, ϕ decreased with increasing W/B ratio. There exists an optimum W/B ratio at which ϕ attains its maximum value and such maximum solid concentration ϕ_{max} of the particle system is taken as the wet packing density. Based on the packing density result, the voids ratio e of the particle system can be determined as: $e = (1 - \phi_{max})/\phi_{max}$.

The voids ratio refers to the ratio of the volume of voids in the bulk volume to the solid volume of the solid particles. Further, the water ratio e_w is defined as the ratio of volume of water to the volume of solid. The excess water ratio e_w' of the cementitious paste can be evaluated as: $e_w' = (e_w - e)$. The excess water ratio has the physical meaning of being the amount of excess water in the paste per solid volume of the particles. Meanwhile, the specific surface area A_S of the particle system is defined as the solid surface area per unit solid volume. Consequently, the WFT can be determined as: WFT = e_w'/A_S.

RESULTS AND DISCUSSIONS

Effects of Superplasticizer

The effects of SP are inspected by referring to the packing density results. From Fig. 2, it can be seen that when the SP dosage was increased from 0% to 0.5%, the packing density of the OPC increased from 0.578 to 0.622 (7.6% increase). Beyond that, the packing density kept increasing with the SP dosage, and eventually attained a value of 0.651 (12.6% increase) and 0.654 (13.1% increase) at a SP dosage of 2% and 3%, respectively. Correspondingly, the voids ratio exhibited a monotonic decrease with increasing SP dosage. The voids ratio at SP dosage of 0%, 0.5%, 1%, 2% and 3% was respectively 0.730, 0.608 (16.8% decrease), 0.577 (20.9% decrease), 0.536 (26.6% decrease) and 0.529 (27.5% decrease). Such packing density improvement and voids ratio reduction can be attributed to the dispersion effect of the SP, which avoids the formation of agglomerates and thus enables the cement grains to be closely packed. However, it is noted that when the SP dosage was higher than 2%, there was little further improvement in packing density. Hence, there exists a saturation dosage for the SP to exert its dispersion effect for improving the particle packing. Such saturation SP dosage was added to the cementitious paste mixes blended with different types of fillers.

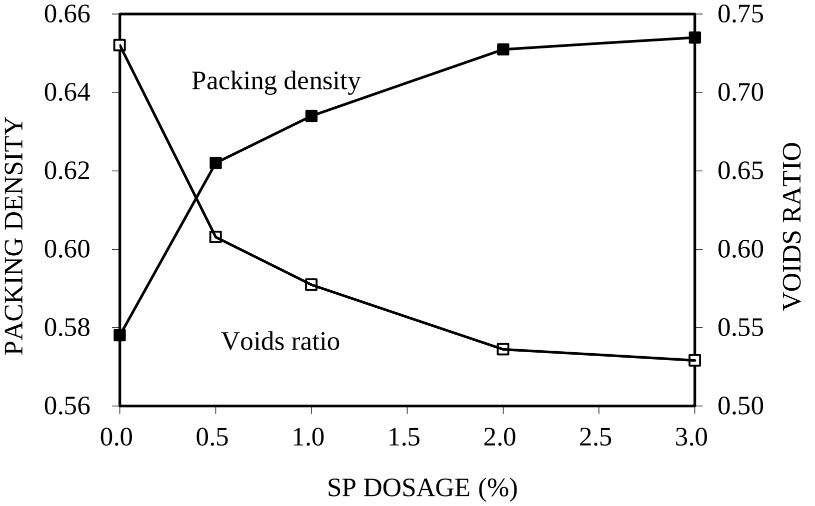

Figure 2   Variations of packing density and voids ratio with SP dosage

Effects of Fillers

The effects of fillers are studied by blending the OPC with SFC, CSF and LSP for packing density measurement. Among all mixes, the amount of SP added was set equal to the saturation dosage, so that the beneficial effect of SP can be kept constant. Fig. 3 shows the packing density results of the (OPC+SFC) blended paste mixes. With 10% SFC added, the packing density increased from 0.640 to 0.676. It continued to increase to 0.692 when the SFC content was 20%. However, further increase in the SFC content resulted in reduction of the packing density to 0.680. Therefore, the packing density did not always increase with the amount of SFC added, and there should be an optimum SFC content for achieving maximum packing density. It should be noted that though the packing density values varied within a small range, the corresponding changes in voids ratio were significant. When the SFC content increased from 0% though 10% to 20%, the voids ratio decreased from 0.563 through 0.479 to 0.445.

Fig. 4 plots the variations of WFT with W/B ratio at different SFC contents. For all SFC contents, the WFT increased linearly with the W/B ratio but at slightly different rates. When the W/B ratio was smaller than or equal to 0.26, the WFT increased with the SFC content. For example, at W/B ratio of 0.20, adding 10% SFC increased the WFT from 0.047 μm to 0.083 μm, while adding 20% SFC further increased the WFT to 0.106 μm. For the range of W/B ratios studied, the sensitivity of WFT to SFC addition decreased at higher W/B ratio. Particularly, at W/B ratios of 0.28 and 0.30, the WFT was respectively 0.30 μm and 0.35 μm approximately, and was not much affected by adding SFC.

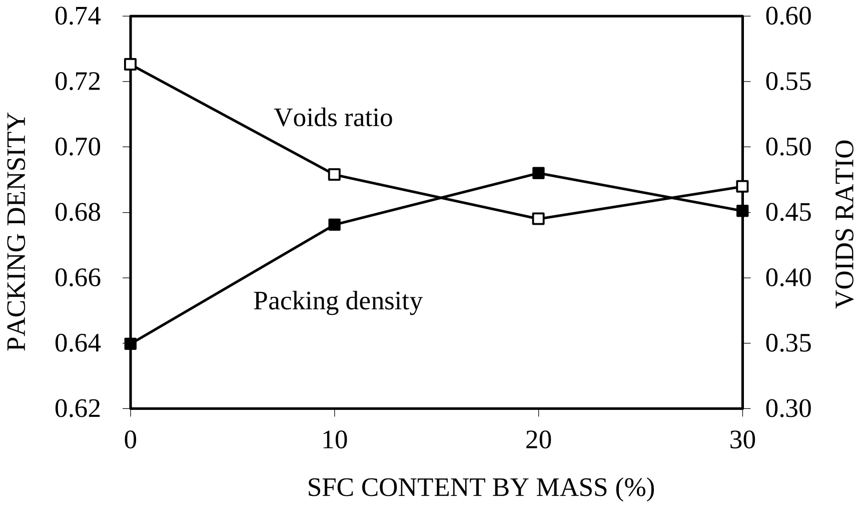

Figure 3 Variations of packing density and voids ratio with SFC content

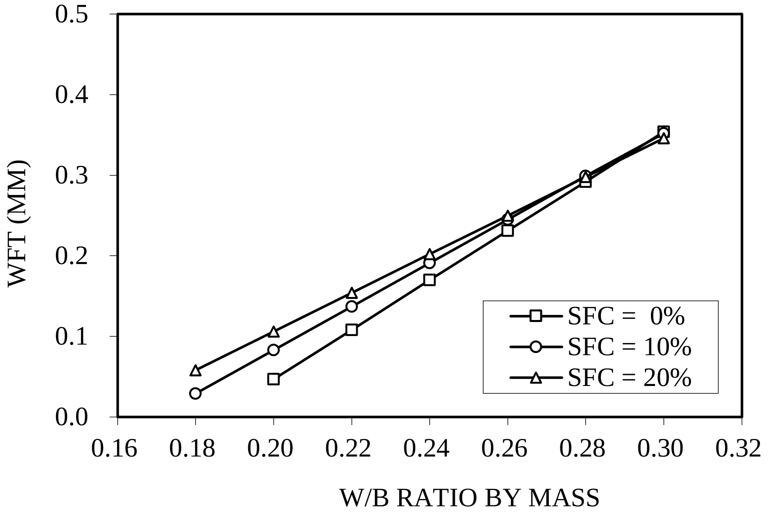

Figure 4 Variation of WFT at different W/B ratio and SFC content

The packing density results of the (OPC+CSF) blended paste mixes are presented in Fig. 5. It can be seen that partially replacing OPC with CSF could significantly improve the packing density. With 5% CSF added, the packing density increased from 0.654 to 0.681 (4.1% increase). With 10% and 15% CSF added, the packing density further increased to 0.707 (8.0% increase) and 0.723 (10.4% increase), respectively. The packing density continued to increase with the CSF content until reaching a value of 0.736 (12.5% increase) at 25% CSF content. However, it is noteworthy that the increase in packing density was generally at a diminishing rate. The voids ratio exhibited a monotonic decrease with increasing CSF content. The voids ratio at CSF content of 0%, 5%, 10%, 15% and 25% was respectively 0.528, 0.468 (11.4% decrease), 0.415 (21.5% decrease), 0.384 (27.3% decrease) and 0.359 (32.1% decrease).

Fig. 6 shows the variations of WFT versus the CSF contents by volume. From the trends of the curves plotted, at W/B ratio of 0.26 and higher, the WFT decreased with increasing CSF content but at different rates. Generally, the rate of decrease slowed down at higher CSF content and lower W/B ratio. Conversely, at W/B ratio of 0.20, the WFT increased slightly as the CSF content increased from 0% to 10%, and then decreased slightly as the CSF content further increased from 10% to 25%. As examples of how the WFT varied with CSF addition, when the CSF content increased from 0% through 10% to 25%, the WFT decreased from 0.338 μm through 0.227 μm to 0.147 μm at W/B ratio equal to 0.32, whereas the corresponding WFT values were 0.051 μm, 0.072 μm and 0.055 μm at W/B ratio equal to 0.20.

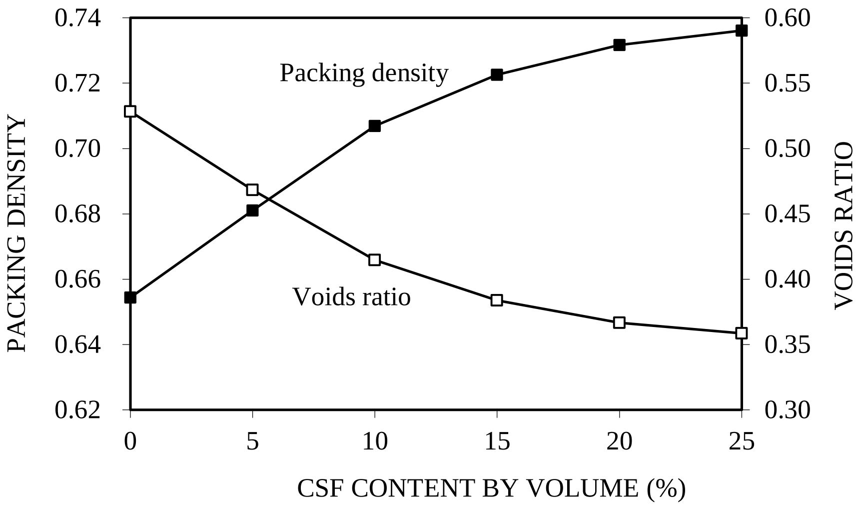

Figure 5 Variations of packing density and voids ratio with CSF content

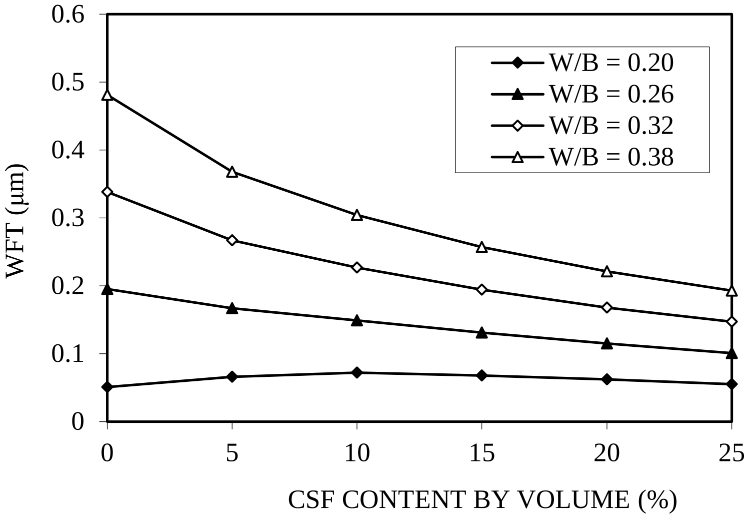

Figure 6 Variation of WFT at different W/B ratio and CSF content

The packing density results of the (OPC+LSP) blended paste mixes are displayed in Fig. 7. It can be seen that in general the packing density increased marginally with the LSP content, the increase was only 2% from 0.640 to 0.653 when 20% LSP content was added. The corresponding decrease in voids ratio was 5.4% from 0.563 to 0.533. Therefore, the incorporation of LSP could slightly improve the packing density and reduce the voids ratio of the paste. Fig. 8 presents the variations of WFT with W/B ratio at different LSP contents. As expected, the WFT increased with the W/B ratio. It can be seen that the addition of LSP reduced the WFT. For example, at W/B ratio of 0.35, the WFT decreased from 0.316 µm to 0.217 µm by adding 20% LSP, whereas at W/B ratio of 0.55, the WFT decreased from 0.742 µm to 0.481 µm by adding 20% LSP.

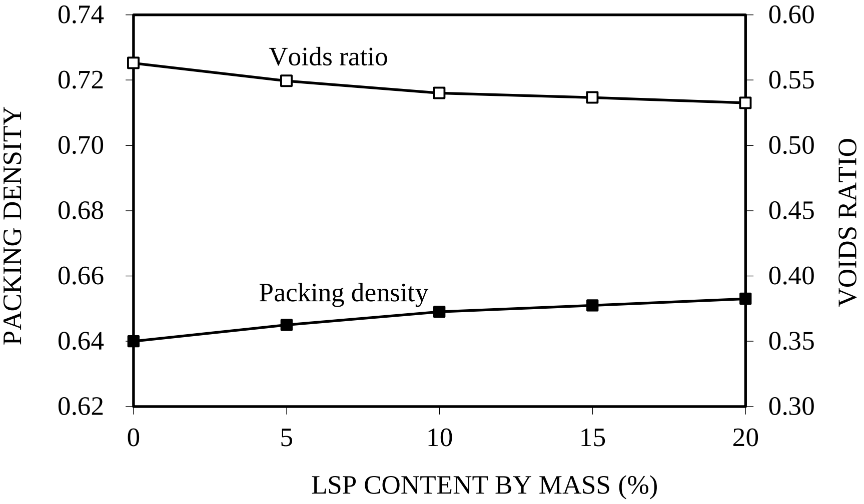

Figure 7 Variations of packing density and voids ratio with LSP content

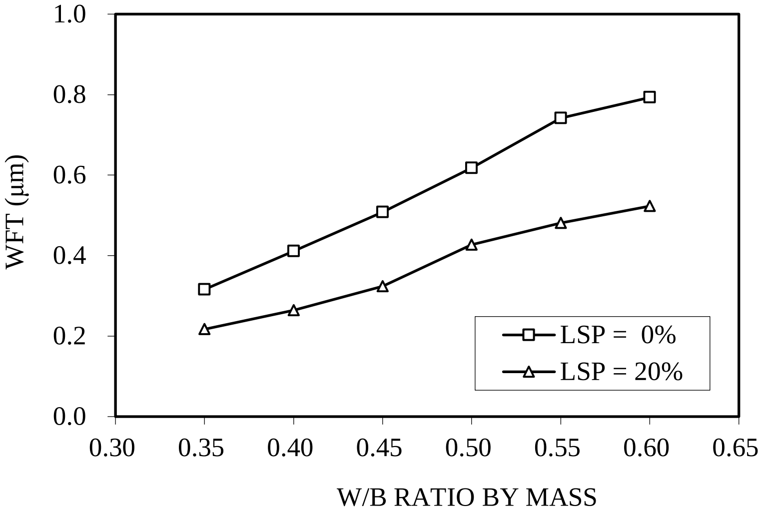

Figure 8 Variation of WFT at different W/B ratio and LSP content

Discussions

The foregoing results of different SP dosages demonstrated the significance of dispersion effect of SP on packing density improvement. With the saturation dosage of SP added, the packing density of the cementitious paste can be further improved by adding micro-fine fillers which fill into the voids between the cement grains. It should be borne in mind that the effectiveness of such filling action could vary greatly among different fillers: replacement of OPC with 20% SFC by mass could notably increase the packing density by 8.1% or reduce the voids ratio by 20.9%; replacement of OPC with 25% CSF by volume could greatly increase the packing density by 12.5% or reduce the voids ratio by 32.1%; addition of 20% LSP to OPC by mass could only slightly increase the packing density by 2.0% or reduce the voids ratio by 5.4%. In other words, the use of SFC and CSF can significantly improve the packing density of binder whereas the use of LSP can only marginally improve the packing density. Such phenomenon can be explained by the particle size of the fillers. Since the SFC and CSF are much finer than the OPC, they can readily fill into the voids between the cement grains and thus effectively improve the packing density. On the contrary, the particle size of

LSP is just slightly finer than OPC, it is too large to fill into the voids between the cement grains and thus its filling effect is relatively small.

It has been reflected from the above that accompanying a small increase in packing density, the corresponding decrease in voids ratio was actually significant. Physically, the water in the cementitious paste has to first fill up the voids between the solid particle skeleton, and only the excess water can play the role to form water films around particles and lubricate the particle system. It should however be noted that the influence on the WFT is two-folded. On one hand, the increased amount of excess water tends to increase the WFT. On the other hand, if the increase in packing density is achieved by addition of fine materials, the corresponding increase in specific surface area tends to decrease the WFT. The overall effect on the WFT is dependent on the relative significance of the above two factors. Generally speaking, by reducing the voids ratio, more excess water will be available for lubrication to enhance the flowability of mortar or concrete at the same W/B ratio. Alternatively, the mortar or concrete strength could be increased through adopting a lower W/B ratio without compromising the flowability. Hence, the combined use of SP and fillers for improving packing density and WFT of the paste phase is advisable in the production of high-performance mortar and concrete.

CONCLUDING REMARKS

In this research, the effects of polycarboxylate-based superplasticizer (SP) and various types of cementitious and inert fillers, namely superfine cement (SFC), condensed silica fume (CSF), and limestone powder (LSP), on the packing density, voids ratio, and water film thickness (WFT) of cementitious paste mixes have been studied. The experimental results have shown that the addition of SP at saturation dosage could improve the packing density by approximately 13% or reduce the voids ratio by 27%. With the saturation dosage of SP added, the addition of SFC, CSF and LSP at contents of 20% by mass, 25% by volume and 20% by mass, could further improve the packing density by approximately 8%, 12% and 2%, and reduce the voids ratio by approximately 21%, 32% and 5% respectively. By comparing the mean particle sizes of SFC, CSF and LSP relative to cement, it has been found that the packing density improvement was more significant for a finer size filler. The variations of WFT at different water/binder (W/B) ratio and different SFC, CSF and LSP contents have been investigated. The results have demonstrated that the WFT always increased with the W/B ratio, and would increase or decrease upon adding fillers depending on the relative significance of increase in excess water and increase in specific surface area. Lastly, the authors remark that the improvement of packing and reduction of voids ratio of cementitious paste would release more excess water for lubricating the paste phase. Hence, the mortar or concrete could have a higher flowability at the same W/B ratio. Alternatively, the mortar or concrete strength could be increased through adopting a lower W/B ratio without compromising the flowability. In view of these benefits, the combined use of SP and fillers to improve the packing density so as to enhance the mortar and concrete performance is recommended.

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