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# Comparative study of effect of raw and densified silica fume in the paste, mortar and concrete



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#### HIGHLIGHTS

• Raw silica fume is better dispersed in paste than densified silica fume.

• Both raw and densified silica fume are good dispersed in concrete.

• Silica fume can significantly improve the properties of ITZ.

• The strength activity index of silica fume in concrete is higher than that in paste.

#### ARTICLE INFO

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#### ABSTRACT

The differences of effect of silica fume in paste, mortar and concrete were studied by determining the non-evaporable water content of pastes, measuring the compressive strengths of pastes, mortars and concretes containing 5% and 10% raw silica fume or densified silica fume with water-to-binder ratios (W/B) of 0.29 and 0.24 and investigating the properties of interfacial transition zone between hardened paste and aggregate. The results show that silica fume can significantly increase the hydration degree of paste. The addition of silica fume trends to increasing the compressive strengths of hardened pastes, mortars and concretes, and the strength activity index of densified silica fume in concrete is the highest while that in paste is the lowest. The agglomeration of silica fume has been found in blended paste which is hardly seen in concrete. The silica fume can improve the interface bond strength between hardened cement paste and aggregate. The crystalline orientation degree, the crystalline size and the content of calcium hydroxide at the interface are obviously decreased by adding silica fume. The different dispersion and the improvement of the interfacial transition zone are the main factors causing the different role of silica fume in paste, mortar and concrete.

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#### 1. Introduction

It is well known that supplementary cementitious materials (SCM) in the form of natural Pozzolan or industrial by-products are able to modify the microstructure and interfacial zones of aggregate-paste or concrete (paste)-reinforcement, and improve durability of concrete. Silica fume (SF) has been recognized as one of the most effective SCMs which contribute to concrete durability improvement through pozzolanic reactions with free lime, pore size refinement and matrix densification, as well as cement paste-aggregate interfacial improvements. SF is a very fine amorphous silica powder produced in electric arc furnaces as a by-product of the manufacture of alloys with silicon or elemental silicon. The typical particle size of silica fume is around

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0.1–0.5  $\mu$ m and the nitrogen BET surface is 20,000 m<sup>2</sup>/kg [1,2]. Numerous studies have proved that the use of SF which is in the nano-range can significantly improve the mechanical and durability properties of Portland cement composite and concrete [3–7]. The high content of amorphous silicon dioxide (more than 85%) and very fine spherical particle are the main reasons for its high pozzolanic reaction [7,8]. The high content of amorphous SiO<sub>2</sub> reacts with calcium hydroxide (CH), which is a weak part of the hydration product of ordinary Portland cement. Furthermore, SF is constituted of smaller particles than cement and can thus fill the gap between cement grains, leading to micro-filling or particle packing which contributes towards an increase of compressive strength and a decreased permeability [7,9,10]. The bondenhancing effect of SF has also been previously discussed in several researches [7,11–14]. The enhanced bond between paste and aggregate or steel rebar has been attributed to both the chemical reaction between CH and pozzolanic materials (pozzolanic



reaction) and the filling effect (physical action) that are considered to significantly densify the interfacial transition zone and reduce the water accumulation beneath rebar and aggregate [15].

However, raw silica fume with particle size distribution smaller than 1  $\mu$ m is rarely used as a SCM due to its low density, making it difficult to handle and transport. Commercial suppliers have responded by processing SF using different methods of densification and compaction in order to improve the handling and transport properties of material, such as densified, slurried or pelletized to increase its bulk density [2]. The densified process of SF promotes the formation of agglomerates with irregular shapes and sizes from 10  $\mu$ m to several hundred microns. It is not easy to disperse the SF particles during the mixing of paste and the agglomerates are not broke down completely due to the high interparticle forces [16,17]. The small particle size and specific surface area partly account for the high activity of SF, and the agglomerates of SF can thus easily reduce its effectiveness on properties of cement paste.

Based on a review of current literatures, it is apparent that the mechanical properties of mortar or concrete mainly depend on the particle size of silica fume and the amount of cement replaced. However, the different effects of silica fume in the paste, mortar and concrete were rarely investigated in the past. Densified silica fume makes different effects on the strength development of paste, mortar and concrete and its performance shown in these systems are also different from that of raw silica fume. In this research, the reaction degree of two kinds of silica fume at different ages was determined. Meanwhile, the compressive strength of paste, mortar and concrete containing two kinds of silica fume was measured as a function of hydrating ages, taking into account of the influence of replacing ratio of silica fume and water to binder ratio. The influence of silica fume on the properties of interfacial transition zone (ITZ) between cement paste and aggregate was specially studied by relevant tests. Then the different effects of silica fume in the paste, mortar and concrete were discussed.

#### 2. Experimental

#### 2.1. Raw materials

P.I 42.5 Portland cement with a specific surface area of 310 m<sup>2</sup>/kg conforming to Chinese National Standard GB 175 and low calcium fly ash conforming to Chinese National Standard GBT1596 were utilized in this study. The chemical compositions of these materials are shown in Table 1.

Two kinds of silica fume were used in this study. Raw silica fume (RSF) with the density of 208 kg/m<sup>3</sup> and densified silica fume (DSF) with the density of 650 kg/m<sup>3</sup> were utilized for preparing test specimens. The chemical compositions of silica fumes are shown in Table 2.

It can be seen from Fig. 1 that the particle size distributions of ultrasonic dispersed densified silica fume are quite similar to that of the raw silica fume. Dry densified silica fume is normally produced by air flotation within silos and the densification process is carried out at temperature much lower than the melting point of silica. Thus, it presumably does not result in any additional permanent bonding between individual spheres, or enlargement of previously formed clusters.

The particle characteristics and morphologies of silica fume were shown in Fig. 2. The agglomerates structure of raw silica fume is loosened with a coarse surface (Fig. 2a) while that of densified silica fume is quite compact with a relatively smooth surface (Fig. 2b) due to the mechanical compression process. However, the particle characteristics of ultrasonic dispersed densified silica fume and raw silica fume are similar to each other (Fig. 2c and d) which agree with the laser diffraction test results as shown in Fig. 1.

#### Table 1

Chemical compositions of cement and fly ash w/%.

#### Table 2

Chemical compositions of silica fumes w/%.

Composition	SiO <sub>2</sub>	K	Na	$H_2O$	LOI
DSF	92.30	0.06	0.02	1.1	2.5
RSF	92.09	0.08	0.02	1.0	2.4

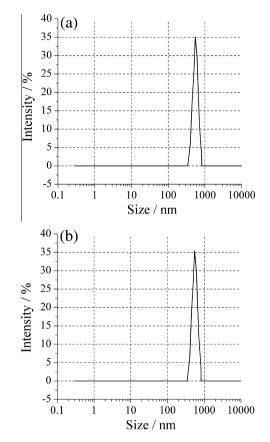


Fig. 1. Particle size distributions of silica fume (a) RSF; (b) DSF.

A polycarboxylate-based superplasticizer was used to adjust the fluidity of mixtures. ISO standard sand conforming to Chinese National Standard GB 17671 was used to prepare mortars. Washed natural river sand and crushed limestone were used as fine and coarse aggregates, respectively. The maximum nominal size of coarse aggregate is 20 mm. The fineness modulus of sand is 2.6.

Mix proportions of pastes, mortars, and concretes are provided in Tables 3–5, respectively. Two series of mixes with water-to-binder ratios (W/B) of 0.29 and 0.24 were prepared. The binder of reference samples contains only cement and fly ash. Mixes containing silica fume were prepared by replacing cement and fly ash with silica fume at levels of 5% and 10% by mass.

#### 2.2. Test methods

Paste cubes of  $40 \times 40 \times 40$  mm, mortar bars of  $40 \times 40 \times 160$  mm, and concrete cubes of  $100 \times 100 \times 100$  mm were prepared. All samples were cured indoor with a constant temperature and humidity ( $20 \pm 1 \circ C$ , over 95% RH) for the initial two days and then, demoulded and placed in water with a constant temperature of  $20 \pm 1 \circ C$ . At the ages of 3, 7, 28, and 90 days, the compressive strengths of all samples and the non-evaporable water content of pastes were tested.

Composition	SiO <sub>2</sub>	$Al_2O_3$	Fe <sub>2</sub> O <sub>3</sub>	CaO	MgO	SO <sub>3</sub>	$Na_2O_{eq}$	LOI
Cement	21.96	4.73	3.68	62.30	2.59	1.82	0.64	1.99
Fly ash	53.33	27.65	6.04	2.86	1.35	0.45	1.54	3.46

*Note:*  $Na_2O_{eq} = Na_2O + 0.658K_2O$ .

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