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THE INFLUENCE OF SILICA FUME ON THE COMPRESSIVE STRENGTH OF CEMENT PASTE AND MORTAR

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ABSTRACT

The compressive strengths of silica fume cement paste and mortar were evaluated at various water-cementitious ratios. Five different water-cementitious ratios were used including, 0.22, 0.25, 0.28, 0.31, and 0.34 and two contents of silica fume, 16% and 25% by weight of cement. Superplasiticizer content was adjusted for each mix to ensure that no segregation would occur.

The results show that the increase in compressive strength of mortar containing silica fume, as a partial replacement for cement, greatly contributes to strengthening the bond between the cement paste and aggregate. Partial replacement of cement by silica fume and the addition of superplasticizer increases the strength of mortar but has no influence on the strength of cement paste. Results were verified by statistical analysis using hypothesis testing at a 95% confidence level.

It was also demonstrated that superplasticizer in combination with silica fume plays a more effective role in mortar mixes than in paste mixes. This can be attributed to a more efficient utilization of superplasticizer in the mortar mixes due to the better dispersion of the silica fume particles. The paper also reviews some of the available literature on the influence of silica fume on cementitious composites and unsettled questions associated with this topic.

Introduction

Silica fume is among one of the most recent pozzolanic materials currently used in concrete. It was first used in 1969 in Norway but only began to be systematically employed in North America and Europe in the early 1980s. Since then, the use of silica fume in concrete has been increasing rapidly; it has been used either as a partial replacement for cement or as an additive when special properties are desired. The rapid increase in the use of silica fume is attributed to its positive effects on the mechanical properties of cementitious composites. Though added strength and low permeability are the two reasons that silica fume is added to concrete, there are other properties that are favorably affected by the addition of silica fume, including: modulus of elasticity [1], drying shrinkage [2, 3], bonding (concrete steel) [4], and resistance to reinforcing steel corrosion and sodium sulfate attack due to low permeability to water and chloride ions [5, 6, 7]. However, some unfavorable properties are associated with the addition of silica fume to concrete, such as loss of slump and reduction in ductility [8].

It is well documented that the use of silica fume as a partial replacement for cement in combination with superplasticizers provides a significant increase in the strength of concrete [9, 10]. The water reduction of fresh concrete and the formation of a more densely compacted matrix at the interfacial zone are believed to be the main causes for this improved durability and strength. However, recent studies have reached contradictory conclusions as to what causes the increase in strength. Some conclude that it is the result of the improved bond strength between the cement paste and the aggregate [11-19], while others conclude that it is mainly due to an increase in the strength of the cement paste matrix [20-24].

Some researchers have demonstrated that the increase in strength with the addition of silica fume to concrete occurs as a result of strengthening the aggregate-matrix transition zone which becomes less porous and more compact in silica fume concrete. Bentur, et al. [11], for example, have reported that the strength of silica fume concrete is greater than that of silica fume paste, which they attribute to the change in the role of aggregate in the concrete. In cement concrete, the aggregate functions as an inert filler but due to the presence of the weak interfacial zone, composite concrete is weaker than cement paste. But, in silica fume concrete, the presence of silica fume eliminates this weak link by strengthening the cement paste-aggregate bond and forming a less porous and more homogeneous microstructure in the interfacial region. Thus, silica fume concrete is stronger than silica fume cement paste, taking into account that the strength of aggregate exceeds the strength of cement paste.

Huang and Feldman [12] have found that mortar without silica fume has a lower strength than cement paste with the same water-cement ratio, while mortar with 30% of the cement replaced by silica fume has a higher strength than cement silica fume paste with the same water-cementitious ratio. They concluded that the addition of silica fume to mortar results in an improved bond between the hydrated cement matrix and the sand in the mix. This improved bond is due to the conversion of the calcium hydroxide, which tends to form on the surface of aggregate particles, into calcium silicate hydrate due to the presence of reactive silica.

Rosenberg and Gaidis [13, 14] have observed no increase in the strength of cement paste at a water-cementitious ratio of 0.24 with the addition of silica fume and superplasticizer, but have observed a substantial increase in the strength of concrete made from the same materials. They reasoned that this increase in the strength of the concrete was due to an improved interfacial bond strength between cement paste and aggregate. Goldman and Bentur [15] have made a similar point by showing that the increase of concrete strength containing silica fume can be attributed to an aggregate-paste bond improvement which is associated with the formation of a less porous transition zone in silica fume concrete.

Baalbake, et al. [16] in their analysis of stress-strain behavior and quantitative microstructural studies have shown the formation of a more compact paste at the interfacial zone in silica fume concrete. This supports the hypothesis that the increase in strength in silica fume concrete should be attributed to the improved bond between cement paste and aggregate. Sarkar and Aitcin [17] have found in their research that there is microstructural evidence of strong bonding between paste and aggregate from a very early age and conclude that this is likely to be an important factor contributing to concrete strength. Larbi [18] has reported that the addition of silica fume increases the compressive strength of mortar significantly. However, silica fume addition does not cause the same increase in strength in cement paste. Based on the results obtained, he concluded that improvements in strength are due to the decrease in thickness of the interfacial zone and better stress transfer between matrix and aggregate particles. Chen and Zhang [19] have reported that the pozzolanic reaction is responsible for the increased strength of the transition zone in silica fume concrete.

In contrast to the studies mentioned above, a number of researchers have reported that the increase in concrete strength is due in great part to a higher quality of the cement paste matrix. Cong, et al. [20], for example, have found that the replacement of cement by silica fume (up to 18%) and the addition of superplasticizer increases the strength of cement paste. Concrete containing silica fume as a partial replacement for cement exhibits an increased compressive strength largely because of the improved strength of the cement paste matrix. But, the changes in the paste-aggregate interface caused by the incorporation of silica fume have little effect on the compressive strength of concrete. Darwin, et al. [21] have found that the strengths of both cement paste and mortar increase when 15% of the cement is replaced by silica fume. However, the ratio of the strength of mortar to the strength of cement paste is lower for the materials containing silica fume which indicates that using silica fume enhances the strength of mortar more than it does that of cement paste. Maher and Darwin [22, 23] have reported from their finite element studies that attaining a perfect bond between cement paste and aggregate provides only an insignificant increase in strength as compared to concrete with normal interfacial strength. Popovics [24] applied various surface treatments to coarse aggregate to increase the compressive strength by increasing the interfacial bond strength. However, in most of his attempts, the surface treatments caused a reduction in compressive strength.

There is basic disagreement among researchers then, as to whether the increase in strength is due to an increase in bond strength between the cement paste and the aggregate by the formation of a more densely compacted matrix at the interfacial zone or is the result of an increase in the strength of the cement paste matrix. The purpose of the research reported here is to improve our understanding of the influence of silica fume on cementitious composites and why the strengths of these composites increase with the addition of silica fume.

Experimental Procedure

Type II Portland cement with an average particle size of 4 μm was used. Silica fume in powder form with an average of 95.75% of SiO_2, and an average particle size of 0.1 μm was used. Its pertinent physical and chemical properties, as provided by the manufacturer, are listed in Table 1. The superplasticizer was a naphthalene formaldehyde sulfonated, manufactured by W.R. Grace, Daracem 100,® with 41% solids content and a relative density of 1.21. The superplasticizer was incorporated in all mixes; the content was adjusted for each mix to ensure that no segregation would occur. For mortar mixes, 1 part cement to 1.4 parts regular concrete sand with a maximum size of 4.76 mm (3/16 in.) was used. The specific gravity of the regular concrete sand was 2.68; fineness modulus was 2.56; and the estimated saturated surface dry condition was 1.8%. The concrete sand was graded and the particle size grading was within the limits of ASTM C33. The additional water was added to the mortar mixes to approach the saturated surface dry condition of the aggregate. Each of the paste and mortar mixes had five different water-cementitious ratios: 0.22, 0.25, 0.28, 0.31, and 0.34 with silica fume contents of 0, 16, and 25 percent by weight of cement.

TABLE 1 - Chemical and Physical Properties of Silica Fume

Chemical composition	Chemical compound	Percent of total weight
	SiO_2	95.75
	Al_2O_3	0.35
	Fe_2O_3	0.21
	CaO	0.17
	MgO	0.09
	SO_3	0.42
	Na_2O	0.51
	Loss of ignition	1.44
Physical properties	Specific gravity	2.25
	Avg. particle size	0.1 microns
	Bulk density	14 lb/ft^3

The cement pastes and mortars were mixed in a Hobart mixer. The mixing procedure was as follows:

1. The cement and silica fume were placed in the mixer. When mixing mortar, the cementitious materials and the sand were placed in the mixer. The mixer was started at medium speed.
2. Half of the mixing water was added and mixing was continued for an additional 2 minutes.
3. The remaining water with the superplasticizer was added and mixing was continued at high speed for 2 more minutes in the case of mortar, and 3 more minutes in the case of paste.
4. The mixture was allowed to rest for 3 minutes, then mixed for an additional minute in the case of mortar and an additional 2 minutes in the case of paste.

The silica fume content was 0%, 16%, and 25% of the weight of cement for all mixtures, whether paste or mortar. Superplasticizer was added as a percentage of the weight of the cementitious materials. The content of superplasticizer was added as follows: 2.5% for mixes of w/c ratios 0.22 and 0.25; 2.0% for mixes of w/c ratio 0.28; 1.5% for mixes of w/c ratio 0.31 and 1.0% for mixes of w/c ratio 0.34. For the silica fume-cement paste mixes with a w/c ratio of 0.22, even with the addition of a relatively large dosage of superplasticizer, the mixtures were stiff and lacked workability; thus;

extra vibration was necessary to mold the specimens. On the other hand, with the same w/c ratio and the same dosage of superplasticizer, the mortar mixes were relatively workable. In all other mixtures, the amount of superplasticizer used was sufficient, and thus, no bleeding or segregation was reported.

Specimens were cast in 51 x 51 x 51 mm (2 x 2 x 2 in.) molds, compacted through external vibration and kept in their molds sealed with plastic sheets for at least 24 hours to prevent moisture evaporation. Specimens with higher dosages of superplasticizer, those of w/c 0.22 and 0.25, exhibited a delay in setting; therefore, these specimens were kept in their molds at least 36 hours before they were placed in the curing room. Specimens were moist cured for 56 days at a temperature of 29 °C (85 °F) and at a relative humidity in excess of 95%. 180 cube specimens were made, six of each mix.

The cubes were tested in compression using a servo-hydraulic controlled testing machine, using spherically seated upper bearing blocks (ASTM C109). No cushioning materials or end caps were used. An initial loading up to one half of the

TABLE 2. Strength of Specimens

Material	Nominal* w/c	Actual w/c	SF** (%)	SP† (%)	Compressive Strength MPa (psi)	Standard Deviation MPa (psi)
Cement paste	0.22	0.235	0	2.50	95.2 (13,800)	8.6 (1,248)
	0.25	0.265	0	2.50	98.6 (14,300)	7.0 (1,012)
	0.28	0.292	0	2.00	93.8 (13,600)	10.5 (1,523)
	0.31	0.319	0	1.50	85.9 (12,450)	5.2 (761)
	0.34	0.346	0	1.00	81.2 (11,780)	8.1 (1,168)
	0.22	0.235	16	2.50	88.1 (12,781)	6.7 (971)
	0.25	0.265	16	2.50	96.5 (13,989)	12.0 (1,740)
	0.28	0.292	16	2.00	93.6 (13,570)	11.0 (1,598)
	0.31	0.319	16	1.50	82.5 (11,968)	5.4 (789)
	0.34	0.346	16	1.00	80.3 (11,640)	8.1 (1,174)
	0.22	0.235	25	2.50	85.3 (12,367)	7.7 (1,115)
	0.25	0.265	25	2.50	98.1 (14,230)	6.7 (974)
	0.28	0.292	25	2.00	89.4 (12,960)	11.8 (1,715)
	0.31	0.319	25	1.50	88.4 (12,825)	9.2 (1,334)
	0.34	0.346	25	1.00	82.6 (11,982)	7.7 (1,121)
Cement mortar	0.22	0.235	0	2.50	93.2 (13,520)	9.2 (1,341)
	0.25	0.265	0	2.50	87.0 (12,609)	8.7 (1262)
	0.28	0.292	0	2.00	83.1 (12,051)	9.6 (1,385)
	0.31	0.319	0	1.50	77.2 (11,190)	6.6 (961)
	0.34	0.346	0	1.00	73.5 (10,661)	7.6 (1,103)
	0.22	0.235	16	2.50	103.1 (14,950)	5.9 (849)
	0.25	0.265	16	2.50	98.8 (14,321)	8.2 (1,189)
	0.28	0.292	16	2.00	94.3 (13,670)	3.9 (571)
	0.31	0.319	16	1.50	84.5 (12,250)	5.8 (837)
	0.34	0.346	16	1.00	82.1 (11,900)	7.2 (1,040)
	0.22	0.235	25	2.50	109.4 (15,860)	6.6 (962)
	0.25	0.265	25	2.50	104.5 (15,158)	7.6 (1,102)
	0.28	0.292	25	2.00	97.8 (14,180)	4.1 (601)
	0.31	0.319	25	1.50	91.4 (13,251)	6.8 (992)
	0.34	0.346	25	1.00	90.1 (13,060)	6.5 (941)

w/c = water-cementitious ratio by weight

* the water in the superplasticizer is not included in the w/c

**SF = silica fume, percent of the cementitious materials

†SP = superplasticizer, is a percent of the cementitious materials

expected maximum loads of the cubes was applied at no specific rate. The rate of loading was adjusted to 241 KPa (35 psi) per second for the remainder of the load, without interruption, up to failure.

Results

Hypothesis testing is used in this study to verify the increase in strength. The theory of a statistical test of hypothesis is to enable the researcher to either reject, or accept the null hypothesis with a measured risk α, which is often referred to the error of estimation. 5% error of estimation or 95% confidence level will be used in this study. More details on this statistical analysis are given elsewhere [25, 26, 27].

Average compressive strengths are summarized in Table 2 and Figures 1 through 3. Each value represents the average of six specimens. The effect of the silica fume content on the strength of cement paste and mortar is shown in Figures 1 and 2.

In the case of cement pastes, the silica fume did not appear to have any effect on strength. At a w/c ratio 0.22, the strength of the SF-cement paste decreased as compared to the control paste; strength was reduced by as much as 8% and 12% with the incorporation of 16% and 25% of silica fume, respectively, as may be seen in Figure 1. This reduction in strength may have resulted from the lack of water in the mix

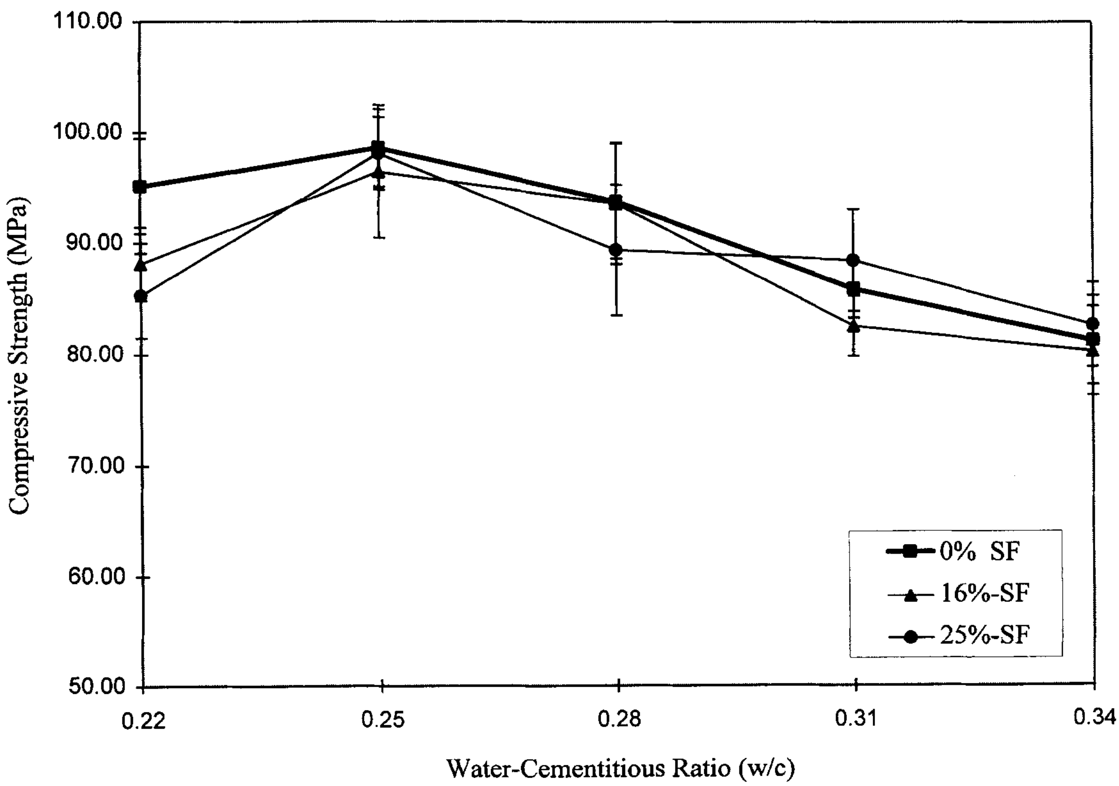

FIG. 1. Compressive strength of cement paste as a function of w/c ratios

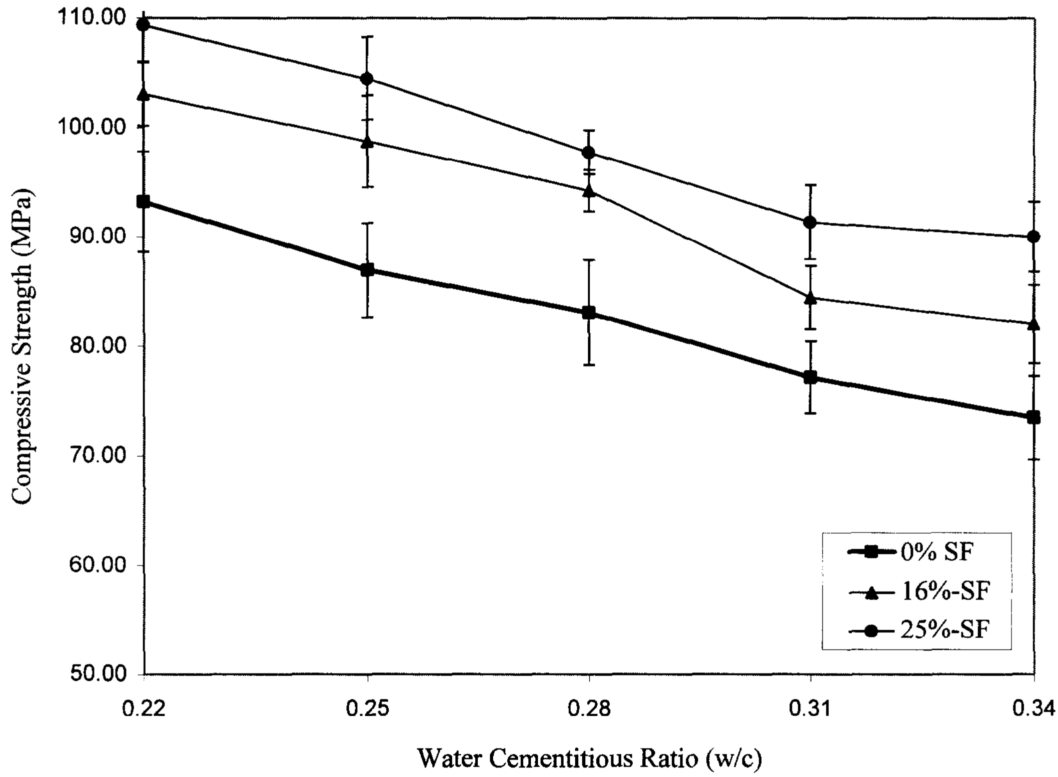

FIG. 2. Compressive strength of mortar as a function of w/c ratios

since self-desiccation of the specimens may have taken place, limiting the pozzolanic effect and thereby reducing the strength of the silica fume cement paste. Although a relatively high dosage of superplasticizer was added to the mixtures - 2.5% by weight of the cementitious materials (which is above the maximum recommended dosage) - they were still stiff and lacked workability. This was due primarily to the high content of silica fume which increased the demand for water in these mixtures. Although the present work did not study the increase of water demand due to the use of silica fume, previous investigations have shown that the demand for water increases as much as 30% depending upon the amount of silica fume in the mixture [5, 23, 28].

In the case of mortars, regardless of the w/c ratios, the strength of the mortar increased with silica fume. Statistical analysis using hypothesis testing at a 95% confidence level indicates a significant increase in strength with silica fume, as may be seen in Figure 2.

Figure 3 compares the strengths of mortar to those of paste with and without silica fume at the five different w/c ratios. It is obvious that the strength of plain cement paste is greater than the strength of plain mortar for all w/c ratios. This is due mainly to the weak interfacial zone between the cement paste and aggregate and the heterogeneous microstructure of concrete. However, with the incorporation of silica fume into the mixture, this is not necessarily true, the strength of SF-mortars was always higher than SF-pastes, regardless of the w/c ratio. The largest difference was

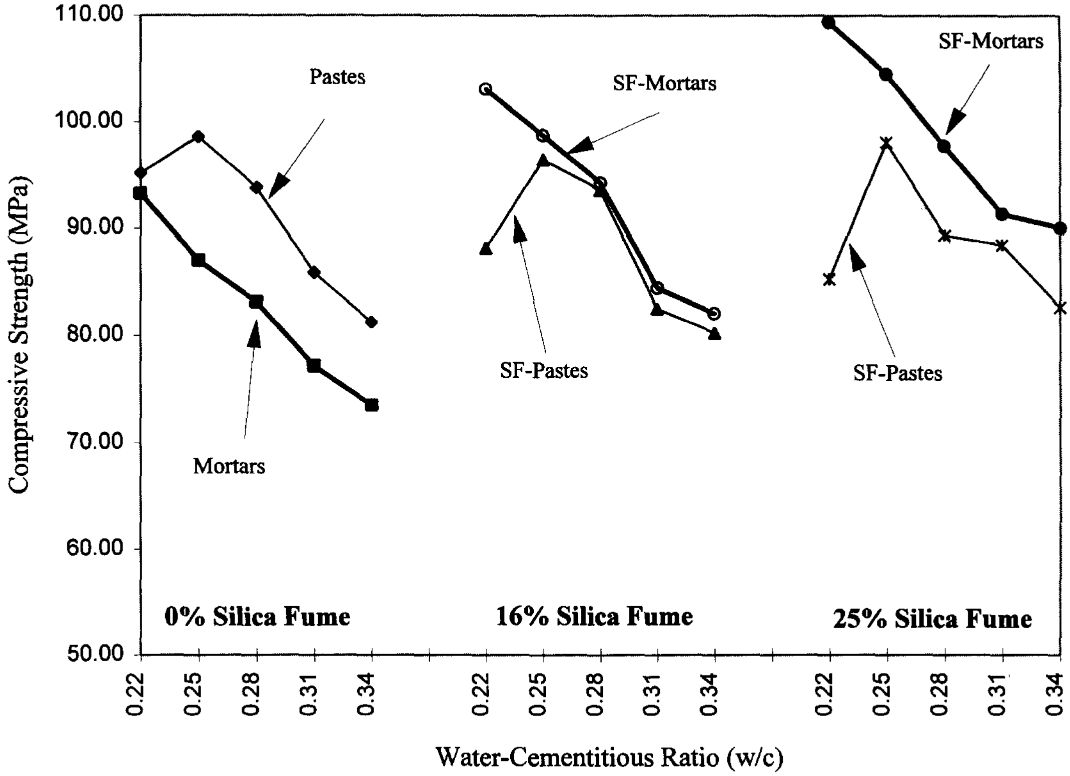

FIG. 3. Cement paste and mortar compressive strengths as a function of w/c ratios

in the case of a w/c ratio of 0.22. At a w/c 0.22 and with a constant dosage of superplasticizer, the cement paste mixes were difficult to mix, with and without silica fume, as compared to silica fume mortar. This was because of the presence of aggregate in the mortar which provided a better mixing action and, thus, better dispersion of the silica fume particles.

It was observed, for the w/c ratios that are used in this study, that at a constant w/c ratio, the dosage of superplasticizer required to obtain an equal workability in SF-cement pastes is not the same as in the SF-mortars. SF-cement paste requires a higher dosage of superplasticizer than SF-mortar to attain proper workability, or a higher content of water. It should be noted that the aggregate used here is in the saturated surface dry condition.

Ratios of mortar strength to paste strength f'_m/f'_p for 0%, 16%, and 25% silica fume contents, at different w/c ratios are compared in Figure 4. Without silica fume, the compressive strength of the paste is greater than the strength of mortar; however, with silica fume, the strength of the paste is less than the strength of the mortar, in which the bond between the aggregate and the cement matrix is much stronger.

Discussion and Conclusions

The optimum benefit of the addition of silica fume is attained when it is used in

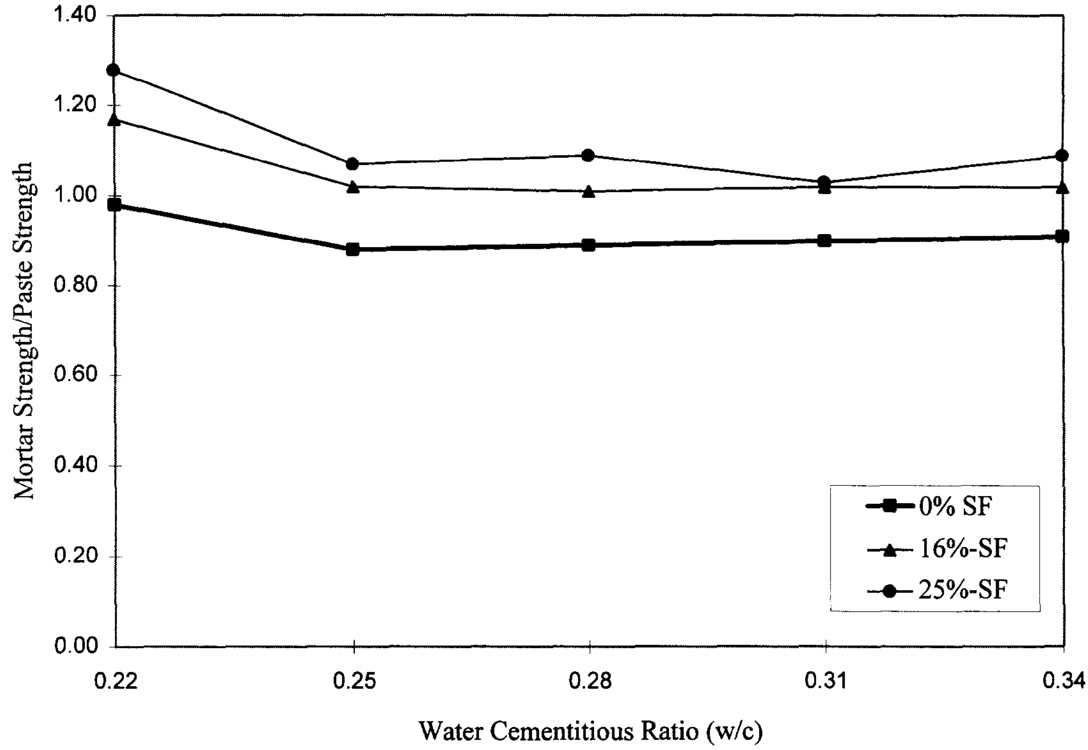

FIG. 4. Ratios of mortar to paste strengths as a function of w/c ratios

combination with superplasticizers. This combination increases the cohesiveness of the fresh composites and reduces the water content. These factors facilitate the formation of a more densely compacted matrix at the interfacial zone which significantly improves the transition zone and, thus, the compressive strength of the system. This may account for the higher strength of silica fume mortars in comparison to pastes.

Some researchers feel that replacing 25% of the cement in composites with silica fume is excessive and that using this much silica fume may actually lower the strength of composites instead of improving it. It has been suggested that the optimum silica fume content should be 15% [29]. This is not supported by the results obtained in this study. The results of this study have shown that 25% silica fume has no adverse effect on strength and, moreover, this percentage cannot be considered the optimum level for maximizing strength. At all w/c ratios between 0.22 and 0.34, the strength of the mortar was found to increase as the silica fume content increased from 16% to 25%. However, having said that, it should be noted that using higher content of silica fume must be accompanied with adjustment to the mix water and the dosage of superplasticizer, to ensure that the specimens will not suffer excessive self desiccation and cracking.

The specific conclusions which can be drawn from the results of this study are as follows:

1. The replacement of cement by silica fume, regardless of the w/c ratio, along with the use of a sufficient amount of superplasticizer, increases the strength of mortar. Strength increases with increasing silica fume content. This can be attributed to the improved aggregate-matrix bond associated with the formation of a less porous interfacial zone and a better interlock between the paste and the aggregate.

2. Silica fume has no strengthening effect on the strength of paste. At the low w/c ratio of 0.22, the SF-cement paste exhibits less strength than the control cement paste; this is due to the lack of water in the mix, resulting in self-desiccation of the specimens.

3. With a constant content of silica fume and a constant dosage of superplasticizer, the optimum strength of SF-cement paste and SF-mortar are reached at different water cementitious ratios.

4. In the cases of the w/c ratios used in this work, at a constant w/c ratio, SF-cement paste requires a higher dosage of superplasticizer than SF-mortar to attain proper workability. The presence of aggregates in the mix seems to provide a better mixing action and thus a better dispersion of the silica fume particles. The aggregate used in this study was in the saturated surface dry condition.

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