



Communication

Improving the workability and strength of silica fume concrete by using silane-treated silica fume

Yunsheng Xu, D.D.L. Chung *

Composite Materials Research Laboratory, State University of New York at Buffalo, Buffalo, NY 14260-4400, USA

Manuscript received 27 October 1998; accepted manuscript 2 December 1998

Abstract

Mortar with high workability even without a water-reducing agent and with high tensile and compressive strengths (3.1 and 78 MPa, respectively) was obtained by using as an admixture silica fume that had been surface treated with a silane coupling agent. The treatment enhanced the wettability of silica fume by water, thereby increasing the workability of the mortar mix. It also enhanced the bond between silica fume and cement, thereby increasing the strength and modulus, both under tension and compression. In particular, the tensile strength was increased by 31% and the compressive strength was increased by 27%. Moreover, the flexural storage modulus, loss tangent, and density were increased. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Silica fume; Workability; Compressive strength; Tensile properties; Cement

The addition of silica fume to concrete is effective for increasing the compressive strength [1–4], decreasing the drying shrinkage [3,4], increasing the abrasion resistance [5], increasing the bond strength with the reinforcing steel [6,7], and decreasing the permeability [8]. As a result, silica fume concrete is increasingly used in civil structures [9,10]. However, silica fume causes workability loss, which is a barrier against proper utilization of silica fume concrete [11]. In this paper, we report that the workability of silica fume mortar is greatly enhanced by using silane-treated silica fume, i.e., silica fume that has been coated with a silane coupling agent prior to incorporation into the mix.

It was recently reported [12] that the surface treatment of silica fume with sulfuric acid prior to incorporation into a cement matrix results in mortars exhibiting increases in tensile strength by 12% and compressive strength by 14%, relative to the values obtained when using as-received silica fume. In this paper, we report that a different surface treatment, which involves the use of a silane coupling agent, provides increases in tensile strength of 31% and compressive strength of 27%, thus resulting in high-strength mortar with tensile strength 3.1 MPa and compressive strength 78 MPa.

The use of silane coupling agents for enhancing the bond between a ceramic filler and a polymer matrix is common

[13] because the epoxy structure at the end of the silane molecule allows coupling with an epoxy matrix. However, silane coupling agents have not been previously used for enhancing the bond between a ceramic filler and a cement matrix. In spite of the difference in chemical structure between cement and polymers, we found that silane coupling agents are effective for both types of matrices. The effectiveness of silane for cement is due to the reactivity of silane's molecular ends with –OH groups and the presence of –OH groups on the surface of both silica and cement.

1. Experimental methods

The cement used was Portland cement (Type I) from the Lafarge Corp. (Southfield, MI). The silica fume (EMS 965, Elkem Materials, Inc., Pittsburgh, PA) was used in the amount of 15% by weight of cement. The silane coupling agent used was a 1:1 (by weight) mixture of Z-6020 ($\text{H}_2\text{NCH}_2\text{CH}_2\text{NHCH}_2\text{CH}_2\text{CH}_2\text{Si}(\text{OCH}_3)_3$) and Z-6040 ($\text{OCH}_2\text{CHCH}_2\text{OCH}_2\text{CH}_2\text{CH}_2\text{Si}(\text{OCH}_3)_3$) from Dow Corning Corp. (Midland, MI). The amine group in Z-6020 serves as a catalyst for the curing of the epoxy and consequently allows the Z-6020 molecule to attach to the epoxy end of the Z-6040 molecule. The trimethylsiloxy ends of the Z-6020 and Z-6040 molecules then connect to the –OH functional group on the surface of the silica fume. The silane was dissolved in ethylacetate. Surface treatment of the silica fume was performed by

* Corresponding author. Tel.: 716-645-2593, ext. 2243; Fax: 716-645-3875; E-mail: ddchung@acsu.buffalo.edu.

immersing it in the silane solution, heating to 75°C while stirring, and holding at 75°C for 1 h, followed by filtration and drying. After this, the silica fume was heated in a furnace at 110°C for 12 h.

The fine aggregate used for mortars was natural sand (all passing #4 US sieve, 99.9% SiO₂). The particle-size analysis of the sand is shown in Fig. 1 of Chen and Chung [14]. No coarse aggregate was used. The sand/cement ratio was 1.00. The water/cement ratio was 0.35. A water-reducing agent (TAMOL SN, Rohm and Haas Co., Philadelphia, PA; sodium salt of a condensed naphthalenesulphonic acid) was used in the amount of 1% of cement weight for mortars with as-received silica fume and 0.2% of cement weight for mortars with treated silica fume, unless stated otherwise. Because the silane treatment greatly enhanced the workability, the need for a water-reducing agent was much decreased. In fact, a water-reducing agent was not needed at all when silane-treated silica fume was used (Table 1). All ingredients were mixed in a rotary mixer with a flat beater. After pouring into molds, an external vibrator was used to facilitate compaction and decrease the amount of air bubbles. The samples were demolded after 24 h and then cured in air—at room temperature and a relative humidity of 100%—for 28 days.

The workability of the mortar mixes was tested by measuring the slump in accordance with the ASTM C143-90a method.

For compressive testing according to ASTM C109-80, specimens were prepared by using a 2 × 2 × 2 inch (5.1 × 5.1 × 5.1 cm) mold. Compression testing was performed using a hydraulic material testing system (MTS, Systems Corp., Eden Prairie, MN). The crosshead speed was 1.27 mm/min. Dog bone-shaped specimens of the dimensions shown in Fig. 1 of Fu et al. [15] were used for tensile testing. They were prepared by using molds of the same shape and size. Tensile testing was performed using a screw-type mechanical testing system (Sintech 2/D, Sintech, Stoughton, MA, USA). The displacement rate was 1.27 mm/min. During compressive or tensile loading up to fracture, the strain was measured by the crosshead displacement in compressive testing or by a strain gauge in tensile testing. Six specimens of each composition were tested.

Dynamic mechanical testing (ASTM D4065-94) at controlled frequencies (0.2, 1.0, and 2.0 Hz) and room temperature (20°C) was conducted under flexure using a Perkin-Elmer Corp. Model DMA 7E dynamic mechanical analyzer.

Measurements of tan δ and storage modulus were made simultaneously as functions of temperature at various constant frequencies. The heating rate was 2°C/min, which was selected to prevent any artificial damping peaks that higher heating rates might cause. The specimens were in the form of beams (120 × 8 × 3 mm) under three-point bending, such that the span was 115 mm. The loads used were all large enough that the amplitude of the specimen deflection was from 6.5 to 9 μm (more than the minimum value of 5 μm that the equipment requires for accurate results). The loads were set so that each type of specimen was always tested at its appropriate stress level. Six specimens of each type were tested.

2. Results

Table 1 shows that the silane treatment causes the silica fume mortar mix to increase in workability (slump). With silane-treated silica fume and no water-reducing agent, the mix's workability is better than it is with as-received silica fume and a water-reducing agent in the amount of 1% by weight of cement. With silane-treated silica fume and a water-reducing agent in the amount of 0.2% by weight of cement, the workability is almost as good as that of the mix with as-received silica fume and a water-reducing agent in the amount of 2% by weight of cement.

Table 2 shows that the silane treatment causes silica fume mortar to increase significantly in tensile strength, tensile modulus, compressive strength, and compressive modulus, in addition to slightly increasing in the tensile and compressive ductilities. The treatment also increased the density. Table 3 shows that the silane treatment causes silica fume mortar to increase greatly in storage modulus, loss modulus, and loss tangent.

3. Discussion

The increase in workability due to silane treatment is because of the improved wettability of silica fume by water after the silane treatment. The improved wettability from the hydrophilic nature of the silane molecule is expected.

The science behind sulfuric acid treatment [12] and silane treatment of silica fume is different. Sulfuric acid treat-

Table 1
Workability of mortar mix

Silica fume	Water-reducing agent/cement	Slump (mm)
As received	0%	150
As received	1%	186
As received	2%	220
Silane treated	0%	194
Silane treated	0.2%	215

Table 2
Tensile and compressive properties and density of mortars with silica fume (with and without silane treatment)

	Without treatment	With treatment
Tensile strength (MPa)	2.35 ± 0.11	3.09 ± 0.13
Tensile modulus (GPa)	10.2 ± 0.07	14.57 ± 0.08
Tensile ductility (%)	0.0141 ± 0.0003	0.0150 ± 0.0004
Compressive strength (MPa)	61.95 ± 2.3	78.4 ± 3.2
Compressive modulus (GPa)	21.08 ± 0.54	28.7 ± 0.46
Compressive ductility (%)	0.138 ± 0.002	0.145 ± 0.003
Density (g/cm ³)*	2.13	2.20

* Measured by using water and Archimedes' principle.

Table 3

Dynamic flexural properties of mortars with silica fume (with and without silane treatment)

	Without treatment	With treatment
Storage modulus (GPa)		
0.2 Hz	20.30 ± 0.02	31.8 ± 0.02
1.0 Hz	20.05 ± 0.02	31.5 ± 0.03
2.0 Hz	19.88 ± 0.02	31.1 ± 0.03
Loss modulus (GPa)*		
0.2 Hz	0.24 ± 0.02	0.74 ± 0.02
1.0 Hz	3 × 10 ⁻⁴	5 × 10 ⁻⁴
2.0 Hz	2 × 10 ⁻⁴	5 × 10 ⁻⁴
Loss tangent		
0.2 Hz	0.0120	0.0234
1.0 Hz	1 × 10 ⁻⁵	2 × 10 ⁻⁵
2.0 Hz	1 × 10 ⁻⁵	2 × 10 ⁻⁵

* Product of storage modulus and loss tangent.

ment roughens the surface of silica fume, as shown by specific surface-area measurement [12]. This is because of the chemical attack of the silica fume by the acid. However, silane treatment involves formation of a silane coating on the surface of the silica fume, so silane treatment does not cause surface roughening. The ability of silane treatment to improve the bond between silica fume and cement matrix (as shown by the improvement in tensile and compressive properties, Table 2) is associated with the chemical coupling—provided by the silane—between silica fume and cement. The coupling results in an increase in density (Table 2), which contributes to causing the increases in tensile and compressive strengths. The increase in storage modulus (Table 3) is consistent with the increases in tensile and compressive moduli (Table 2). The increase in loss tangent (Table 3) is probably due to the contribution of the silane coating to viscoelastic damping.

Compared with the sulfuric acid treatment, silane treatment is less hazardous and of less concern to the pH of the concrete. The pH affects the corrosion resistance of steel reinforcement in concrete.

4. Conclusion

The surface treatment of silica fume with a silane coupling agent prior to incorporation into mortar was found to greatly enhance the workability of the silica fume mortar mix and cause the tensile strength to increase by 31% and the compressive strength to increase by 27%, relative to the

values obtained without treatment. The effect on workability is due to the enhanced wettability of silica fume by water. The effect on strength is due to the improved bond between silica fume and cement and to the increased density of the mortar. In addition, the silane treatment resulted in increases in tensile and compressive moduli, and in flexural storage modulus and loss tangent.

Acknowledgments

The authors thank Dr. Lin Li of the Perkin-Elmer Corp. (Norwalk, CT) for loaning the fixture for dynamic flexural property testing.

References

- [1] B.B. Sabir, High-Strength Condensed Silica Fume Concrete, *Mag Concr Res* 47 (1995) 219–226.
- [2] H.A. Toutanji, T. El-Korchi, Tensile and Compressive Strength of Silica Fume-Cement Pastes and Mortars, *Cem Concr Aggr* 18 (1996) 78–84.
- [3] M.N. Haque, Strength Development and Drying Shrinkage of High-Strength Concretes, *Cem Concr Compos* 18 (1996) 333–342.
- [4] Pu-Woei Chen, D.D.L. Chung, Concrete Reinforced with up to 0.2 vol.% of Short Carbon Fibers, *Composites* 24 (1) (1993) 33–52.
- [5] Zeng-Qiang Shi, D.D.L. Chung, Improving the Abrasion Resistance of Mortar by Adding Latex and Carbon Fibers, *Cem Concr Res* 27 (8) (1997) 1149–1153.
- [6] T.A. Bürge, Densified Cement Matrix Improves Bond with Reinforcing Steel, in: P. Bartos (Ed.), *Bond in Concrete*, Applied Science Publishers, London, 1982, pp. 273–281.
- [7] O.E. Gjorv, P.J.M. Monteiro, P.K. Mehta, Effect of Condensed Silica Fume on the Steel-Concrete Bond, *ACI Mater J* 87 (1990) 573–580.
- [8] J.G. Cabrera, P.A. Claisse, Measurement of Chloride Penetration into Silica Fume Concrete, *Cem Concr Compos* 12 (1990) 157–161.
- [9] M.D. Luther, Silica-Fume (Microsilica) Concrete in Bridges in the United States, *Trans Res Rec* 1204 (1988) 11–20.
- [10] A.A. Bubshait, B.M. Tahir, M.O. Jannadi, Use of Microsilica in Concrete Construction, *Building Research Information* 24 (1) (1996) 41–49.
- [11] J. Punkki, J. Golaszewski, O.E. Gjorv, Workability Loss of High-Strength Concrete, *ACI Mater J* 93 (5) (1996) 427–431.
- [12] Xiaohui Li, D.D.L. Chung, Improving Silica Fume for Concrete by Surface Treatment, *Cem Concr Res* 28 (4) (1998) 493–498.
- [13] Y. Xu, D.D.L. Chung, Increasing the Thermal Conductivity of Boron Nitride and Aluminum Nitride Particle Epoxy-Matrix Composites by Particle Surface Treatments, *J Electron Packaging*, in press.
- [14] Pu-Woei Chen, D.D.L. Chung, Carbon Fiber Reinforced Concrete as a Smart Material Capable of Non-Destructive Flaw Detection, *Smart Mater Struct* 2 (1993) 22–30.
- [15] Xuli Fu, Weiming Lu, D.D.L. Chung, Improving the Tensile Properties of Carbon Fiber Reinforced Cement by Ozone Treatment of the Fiber, *Cem Concr Res* 26 (10) (1996) 1485–1488.