Pergamon

Cement and Concrete Research, Vol. 28, No. 4, pp. 493–498, 1998

0008-8846/98 $19.00 + .00

PII S0008-8846(98)00017-9

IMPROVING SILICA FUME FOR CONCRETE BY SURFACE TREATMENT

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(Received September 23, 1997; in final form January 16, 1998)

ABSTRACT

Surface treatment of silica fume with sulfuric acid prior to incorporation in a cement matrix resulted in composites exhibiting increases in tensile strength by 12%, tensile modulus by 72%, tensile ductility by 57%, abrasion resistance by 20%, loss tangent by 30–80%, flexural storage modulus by 80–120%, and flexural loss modulus by 160–300%, relative to the values obtained by using as-received silica fume. © *1998 Elsevier Science Ltd*

Introduction

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).

It is well known that surface treatment of a filler (or a reinforcement) can be effective for increasing the bond strength between the filler and the matrix, thereby improving the properties of the composite. However, surface treatment of silica fume has not been previously investigated. In this paper, we report that surface treatment of silica fume greatly improves the properties of the resulting silica fume concrete (actually mortar and cement paste). The properties addressed are the tensile and compressive properties of mortars, the abrasion resistance of mortars, and the dynamic flexural properties (relevant to vibration damping) of cement pastes.

Experimental Methods

The cement used was Portland cement (Type I) from Lafarge Corp. (Southfield, MI). The silica fume (Elkem Materials, Inc., Pittsburgh, PA, EMS 965) was used in the amount of 15% by weight of cement.

Surface treatment of silica fume was performed by immersing the silica fume in sulfuric acid (96%) for 2 h, washing with water, filtering, and then drying at 150°C for 1–2 days. The

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Communicated by D.M. Roy.

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effect of the treatment on the BET specific surface area was investigated by nitrogen adsorption and measurement of the pressure of the gas during adsorption using the Micromeritics ASAP 2010 instrument. The effect of the treatment on the morphology was investigated by scanning electron microscopy.

The fine aggregate used for mortars was natural sand (all passing #4 US sieve, 99.9% SiO_2); the particle size analysis of the sand is shown in Figure 1 of Ref. 10; no coarse aggregate was used; the sand/cement ratio was 1.5 for mortars for abrasion testing and 1.0 for mortars for tensile and compressive testing. The water/cement ratio was 0.45 for mortars for abrasion testing, 0.35 for mortars for tensile and compressive testing, and 0.35 for cement pastes for dynamic flexural testing. 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 2% of cement weight for mortars and in the amount of 3% of cement weight for cement pastes. All ingredients were mixed in a Hobart mixer with a flat beater. After pouring into square 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 40% for 28 days.

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). The cross head speed was 1.27 mm/min. Dog-bone shaped specimens of the dimensions shown in Figure 1 of Ref. 11 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). The displacement rate was 1.27 mm/min. During compressive or tensile loading up to fracture, the strain was measured by the cross-head displacement in compressive testing or by a strain gage in tensile testing. Six specimens of each composition were tested.

The abrasion resistance was measured using ASTM C944–90a (Rotating-Cutter Method). The rotating cutter involved 24 No. 1 Desmond-Huntington grinding dressing wheels (diameter = 1.5 inch, thickness = 3/32 inch) and washers (diameter = 1 3/8 inch, thickness = 1/32 inch) between the dressing wheels (Fig. 1 of Ref. 5). The total cutting area was 71.12 cm^2. The load was 10 kgf (98 N) during abrasion. The speed of the rotating cutter was 200 rpm. Each specimen was abraded once on each of its two opposite flat sides, such that each time lasted 2 min. The weight after each time was measured using a balance to an accuracy of 0.1 g. Six specimens of each type of mortar were tested. The average weight loss was used to compute the depth of wear, using the separately measured density of each mortar.

Dynamic mechanical testing (ASTM D4065–94) at controlled frequencies (0.20, 1.00, and 2.00 Hz) and temperatures (30–150°C) were 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 a function of temperature at various constant frequencies. The heating rate was 2°C/min, which was selected to prevent any artificial damping peaks which may be caused by higher heating rates. The specimens were in the form of beams (24 × 8 × 3 mm) under three-point bending, such that the span was 20 mm. The loads used were all large enough so that the amplitude of the specimen deflection was always over the minimum value of 5 μm required by the equipment for accurate results. The loads were set so that each different type of specimen was always tested at its appropriate stress level. At least seven specimens of each type were tested.

TABLE 1
Effect of surface treatment of silica fume on tensile and compressive properties of mortar.

	As-received	Surface treated
Tensile strength (MPa)	2.38 ± 0.08	2.67 ± 0.08
Tensile modulus (GPa)	9.80 ± 0.06	16.9 ± 0.5
Tensile ductility (%)	0.0051 ± 0.0007	0.0080 ± 0.0003
Compressive strength (MPa)	31.3 ± 1.9	35.6 ± 0.9
Compressive modulus (GPa)	20.9 ± 2.5	21.0 ± 2.1
Compressive ductility (%)	0.15 ± 0.02	0.17 ± 0.02

Results and Discussion

The specific surface area of silica fume was 17 and 23 m^2/g before and after surface treatment respectively. Microscopy showed no change in morphology after surface treatment.

Table 1 shows that the tensile strength, modulus and ductility, and the compressive strength were all increased by the surface treatment. The compressive modulus and ductility were not changed by the surface treatment. The greatest effects pertained to the tensile modulus and ductility.

The abrasion resistance was improved by the surface treatment of the silica fume, as shown by the decrease in the wear depth, whether for the top face (top side of specimen in the mold) or the bottom face (Table 2). For each specimen, the abrasion resistance was better for the top face than the bottom face, due to the tendency for silica fume (lightweight) to float in the

TABLE 2
Effect of surface treatment of silica fume on abrasion resistance of mortar.

		Wear depth (mm)	
Sample	Density (g/cm^3)	Top face	Bottom face
As received, #1	2.01	0.147	0.158
As received, #2	2.02	0.146	0.157
As received, #3	2.05	0.134	0.148
As received, #4	2.04	0.140	0.151
As received, #5	2.03	0.144	0.150
As received, #6	2.05	0.139	0.147
As received, average	2.03	0.142	0.152
Treated, #1	2.07	0.111	0.121
Treated, #2	2.06	0.114	0.124
Treated, #3	2.04	0.118	0.127
Treated, #4	2.07	0.113	0.120
Treated, #5	2.05	0.116	0.124
Treated, #6	2.09	0.109	0.118
Treated, average	2.06	0.114	0.122

mortar mix in the mold. In other words, the concentration of silica fume decreased from the top face to the bottom face.

The loss tangent, storage modulus, and loss modulus at all temperatures and frequencies studied were all increased by surface treatment of the silica fume (Table 3). The storage modulus increased by up to 120% after the treatment; the loss tangent increased by up to 80% after the treatment; the loss modulus increased by up to 300% after the treatment. The effect on the storage modulus tended to decrease with increasing temperature, though the effect was still large (83–88% increase) at the highest temperature of 150°C. The loss tangent decreased with increasing frequency at any temperature; the decrease was particularly large when the frequency was increased from 1.0 to 2.0 Hz. A similar dependence on frequency occurred for the loss modulus. The storage modulus did not vary monotonically with frequency; the highest storage modulus was observed at 1.0 Hz at any temperature.

Discussion

All the effects reported here concerning surface treatment of silica fume are desirable for practical applications. These effects include increases in the tensile strength, modulus and ductility, abrasion resistance, loss tangent (which describes the vibration damping capacity), and storage modulus (which describes the stiffness). All these effects, except that concerning the loss tangent, are attributed to the improved bonding between silica fume and cement matrix after the surface treatment. The origin of the improved bonding has not been investigated, though it may be related to the change in surface functional groups. The increase in loss tangent due to the surface treatment is at least partly due to the increase in interface area resulted from the increase in specific surface area of the silica fume.

The storage modulus attained by using surface-treated silica fume is much higher than the value previously reported for cement paste containing as-received silica fume together with methylcellulose (12), which is comparable to the value reported here for cement paste containing as-received silica fume only and is higher than the value for cement paste containing latex (12). The loss tangent attained by using surface-treated silica fume at 0.2 and 1.0 Hz is higher than the value previously reported for cement paste containing latex, which is in turn higher than that for cement paste containing as-received silica fume together with methylcellulose (12). The loss modulus attained by using surface treated silica fume at 0.2 or 1.0 Hz is much higher than that previously reported for cement paste containing as-received silica fume together with methylcellulose, which is in turn higher than that for cement paste containing latex (12). Hence, surface-treated silica fume is more effective than latex for vibration damping.

The high static tensile modulus attained by using surface-treated silica fume is consistent with the high dynamic flexural storage modulus. Both properties describe the stiffness. They are also consistent with the high abrasion resistance. The high tensile ductility attained by using surface treated silica fume is consistent with the high vibration damping ability.

The surface treatment of the silica fume affected the tensile properties much more than the compressive properties. This is because the tensile properties are more dependent on the filler-matrix bond strength than the compressive properties.

TABLE 3
Effect of surface treatment of silica fume on dynamic flexural properties of cement paste.

	As received	Treated
Loss tangent		
30°C, 0.2 Hz	0.112	0.172
30°C, 1.0 Hz	0.051	0.091
30°C, 2.0 Hz	<0.0001	<0.0001
60°C, 0.2 Hz	0.103	0.161
60°C, 1.0 Hz	0.054	0.093
60°C, 2.0 Hz	<0.0001	<0.0001
90°C, 0.2 Hz	0.110	0.182
90°C, 1.0 Hz	0.064	0.084
90°C, 2.0 Hz	<0.0001	<0.0001
120°C, 0.2 Hz	0.109	0.175
120°C, 1.0 Hz	0.061	0.091
120°C, 2.0 Hz	<0.0001	0.0001
150°C, 0.2 Hz	0.108	0.168
150°C, 1.0 Hz	0.055	0.087
150°C, 2.0 Hz	<0.0001	<0.0001
Storage modulus (GPa, ±0.02)		
30°C, 0.2 Hz	5.46	11.29
30°C, 1.0 Hz	6.85	15.02
30°C, 2.0 Hz	6.18	11.65
60°C, 0.2 Hz	5.90	11.92
60°C, 1.0 Hz	7.08	14.52
60°C, 2.0 Hz	6.27	11.74
90°C, 0.2 Hz	5.82	12.28
90°C, 1.0 Hz	7.05	13.67
90°C, 2.0 Hz	6.16	11.57
120°C, 0.2 Hz	5.68	10.99
120°C, 1.0 Hz	6.86	13.30
120°C, 2.0 Hz	6.07	11.24
150°C, 0.2 Hz	5.72	10.78
150°C, 1.0 Hz	6.74	12.66
150°C, 2.0 Hz	6.03	11.06
Loss modulus (GPa, ±0.02) (product of loss tangent and storage modulus)		
30°C, 0.2 Hz	0.61	1.94
30°C, 1.0 Hz	0.34	1.37
30°C, 2.0 Hz	<0.0006	<0.001
60°C, 0.2 Hz	0.61	1.92
60°C, 1.0 Hz	0.38	1.35
60°C, 2.0 Hz	<0.006	<0.001
90°C, 0.2 Hz	0.64	2.23
90°C, 1.0 Hz	0.45	1.15
90°C, 2.0 Hz	<0.0006	<0.001
120°C, 0.2 Hz	0.62	1.92
120°C, 1.0 Hz	0.42	1.21
120°C, 2.0 Hz	<0.0006	<0.001
150°C, 0.2 Hz	0.62	1.81
150°C, 1.0 Hz	0.37	1.10
150°C, 2.0 Hz	<0.0006	<0.001

Conclusion

Surface treatment of silica fume with sulfuric acid prior to incorporation of silica fume in a cement matrix was found to increase the tensile strength (by 12%), modulus (by 72%) and ductility (by 57%), abrasion resistance (by 20%), loss tangent (by 30–80%) and dynamic flexural storage modulus (by 80–120%), and loss modulus (by 160–300%) of cement paste or mortar, relative to the values obtained by using as-received silica fume. These effects are probably due to increased bond strength between silica fume and cement matrix and increased specific surface area of the silica fume.

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