EXAMINATION OF STRUCTURAL PROPERTIES OF POLY VINYL ALCOHOL AND ITS SIGNIFICANCE

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ABSTRACT

The structure and properties of mortars and concretes containing up to 2 wt% (based on cement) of poly(vinyl alcohol) (PVA) were examined and compared with those without PVA. Among changes occurring with the addition of PVA were increases in air void content and apparent fluidity and a reduction in the bleeding of fresh mortar and concrete. The increased fluidity caused increased slump for fresh concrete. The microstructure was examined by polarizing optical microscopy and scanning electron microscopy in backscattered mode of cut surfaces after hardening. The porous interfacial transition zones around sand grains and coarse aggregate were significantly reduced, and the cement particles were uniformly distributed without significant depletion near aggregate surfaces. For mortars, using a prewetting mixing technique, the compressive strength was decreased moderately, but the flexural strength was unchanged. For concretes, with the same mixing technique, the compressive strengths after 28 days of hydration were relatively unchanged, but the postpeak area of the compression stress-strain curve was reduced, accompanying a change in fracture behavior from debonding to cohesive failure of the coarse aggregate.

I INTRODUCTION

The strength of concrete is known to increase with an in-crease in the aggregate-cement bond strength [1–5]. Both silica fume [1] and polymer dispersions [2–5] have been used to increase concrete strength by improving the aggre-gate-cement bond. However, these materials need to be used at the rate of 10–

20% based on cement to be effective, and at this rate, they can add considerably to the cost. Small amounts of poly (vinyl alcohol) (PVA) have been shown to significantly increase the aggregate-paste bond strength and reduce the thickness of the interfacial transition zone in an aggregate-paste bond model system [6,7]. If the increase in bond strength with PVA observed with this model system occurs in concrete as well, an increase in the strength of the concrete can be expected. On the other hand, water soluble polymers like PVA have often been found to decrease the mechanical properties of mortar and concrete. In addition to affecting the bond strength, PVA has long been known to affect the air content of paste and mortar. In the present study, the combined effects arising from PVA of paste-aggregate bond improvement and increased air con-tent were examined with regard to the strength properties of mortar and concrete.

II EXPERIMENTAL

2.1. Materials

The cement used was a commercial ASTM type I ordinary port-land cement. The two sands used were ASTM graded sand (silica sand, ASTM C778, U.S. Silica Co., Ottawa, IL, USA, used to prepare mortars and vacuum-cast concretes, and river sand with a maximum grain size of 4 mm, used to prepare regularly cast concretes. Pea gravel with the size range of 6–13 mm (1/4–1/2 in) was used as the coarse aggregate for the concrete. To remove clay from the aggregate surface, the pea gravel was cleaned with silica sand and water in a drum mixer and dried in air. The PVA used was Airvol 805 (Air Products and Chemicals Inc. Allentown, PA, USA). This had a relatively low molar mass of 31,000–50,000 weight average, 15,000–27,000 number average, and was 87–89% hydrolyzed PVA. Airvol 805 is a low foaming grade. PVA powder was dispersed in cold water at a concentration of 20%, and this mixture was heated to 908C with stirring to completely dissolve the PVA. Then the PVA solution was cooled to room temperature and used within 24 h.

2.2. Mixing, casting, and curing procedures

For the mortars, a cement: sand: water ratio of 1:2:0.5 was used. When PVA was added, the amount of water in the PVA solution was included in the water-to-cement ratio. The amounts of PVA added were 1% and 2% (solid) based on the weight of cement; these corresponded to 0.29 and 0.57 wt% based on the total weight of mortar. ASTM C305 mixing was used as the standard mixing procedure for the mortar, using a planetary mixer (Hobart Co., Troy, OH, USA). ASTM C305 mixing consists of a sequence of mixings that involve a total of 1.0 min at a paddle speed of 140 rpm followed by a total of 1.5 min at a speed of 285 rpm. The reference mortar was mixed following the ASTM C305 mixing procedure. When PVA was added, the prewetting technique described previously [8] was employed to minimize air void formation. In this technique cement, sand, and water were first mixed using the ASTM C305 procedure, and then the PVA solution was mixed with the prewetted mortar at a paddle speed of 140 rpm for 1 min. The fresh mortar was cast into plastic molds on a vibrating table. Plastic cylinder molds 76 mm (3 in) in diameter and 152 mm (6 in) high were used for compression tests, and square-bar molds 51 3 51 3 305 mm (2 3 2 3 12 in) were used for flexural strength tests. The volumes of each mold and the weights of the mortar in the molds were measured to deter-mine the density of the mixtures.

For the concretes, a cement:sand:pea gravel ratio of 1:2:2.5 and two water-to-cement ratios (0.50 and 0.45) were used. The amounts of PVA added were again 1% and 2% (solids), based on the weight of cement. These correspond to 0.17% and 0.33% based on the total weight of concrete with a water-to-cement ratio of 0.50. The amounts of water in the PVA solution were included in the water-to-cement ratios. An omnimixer (Chiyoda Technical and Industrial Co., To-kyo, Japan) was used for concrete mixing. This had a moving bowl with a rubber wall and a mixing rod in the middle. To make the reference concretes, cement, sand, pea gravel, and water were mixed together for 2 min at a speed of around 350 rpm and then for another 1 min at a speed of around 250 rpm. To make the PVA-modified concretes, cement, sand, pea gravel, and water were first mixed for 2 min at 350 rpm, and then the PVA solution was mixed with the prewetted concrete for 1 min at 250 rpm to minimize air void formation [8]. The fresh concretes were cast into plastic cylinder molds 102 mm (4 in) in

diameter and 203 mm (8 in) high with vibration for compression testing. The volumes of each molds and the weights of the concrete in the molds were measured to deter-mine the densities of the fresh mixtures.

2.3. Mechanical strength tests

The compressive strength of cylindrical specimens of mortar and concrete were measured with a hydraulic Instron Model 1336 (Instron Corp., Canton, MA, USA) equipped with a 2225 kN (500 kips) load-cell at a crosshead speed of 0.025 mm/s (0.001 in/sec). Four cylinder specimens, 76 mm (3 in) in diameter and 152 mm (6 in) high for mortar and 102 mm (4 in) in diameter and 203 mm (8 in) high for con-crete, were tested for each mix. The load and machine dis-placement signals were recorded using a data acquisition system. Two hour before testing, the hardened specimens were taken out of the water bath where they had been for 7 or 28 days to dry the surfaces and cap both ends of the cyl-inder with sulfur mortar (ASTM C617).

The flexural strength of the square mortar bars, with the dimensions of 51 3 51 3 305 mm (2 3 2 3 12 in), was measured by four-point bending with a span length of 229 (9 in) using an Instron Model 4206 (Instron Corp., Canton, MA, USA) equipped with a 44.5-kN (10 kips) load-cell at a crosshead speed of 0.0212 mm/s (0.05 in/min). The specimens were taken out of the water bath just before testing and their surfaces were maintained wet during the test to prevent surface drying cracks. Six specimens from each mix were tested.

2.4. Observation of interfacial transition zone and fracture surface

To examine the microstructure, the tested mortars and concretes were sectioned with a diamond saw, and their cut surfaces were observed with a polarizing optical micro-scope (Olympus BH-2, Tokyo, Japan) and a scanning electron microscope in backscatter mode. For backscatter microscopy, the sectioned pieces were immersed in a low viscosity epoxy resin after drying in air to allow the specimens to become impregnated with the resin. The resin was then hardened at room temperature for one day, and the specimen surface was polished with #1000, #2400, and #4000 silicon carbide grinding papers. The nonpolar liquid (kerosene) was used as a coolant for the polishing and for washing the specimens in an ultrasonic cleaner between polishing steps. The polished specimens were dried under vacuum for two days and thinly coated with Au-Pd. The specimens were examined with a scanning electron micro-scope (Hitachi S-520, Hitachi Ltd., Tokyo, Japan) equipped with a backscattered electron detector.

The fracture surfaces of the tested concrete specimens were also observed visually and with an optical stereo microscope.

III RESULTS

3.1 Mortar

3.1.1. Fresh mortar properties

The addition of PVA to mortar was found to cause the mass density to decrease, and the decrease was greater with
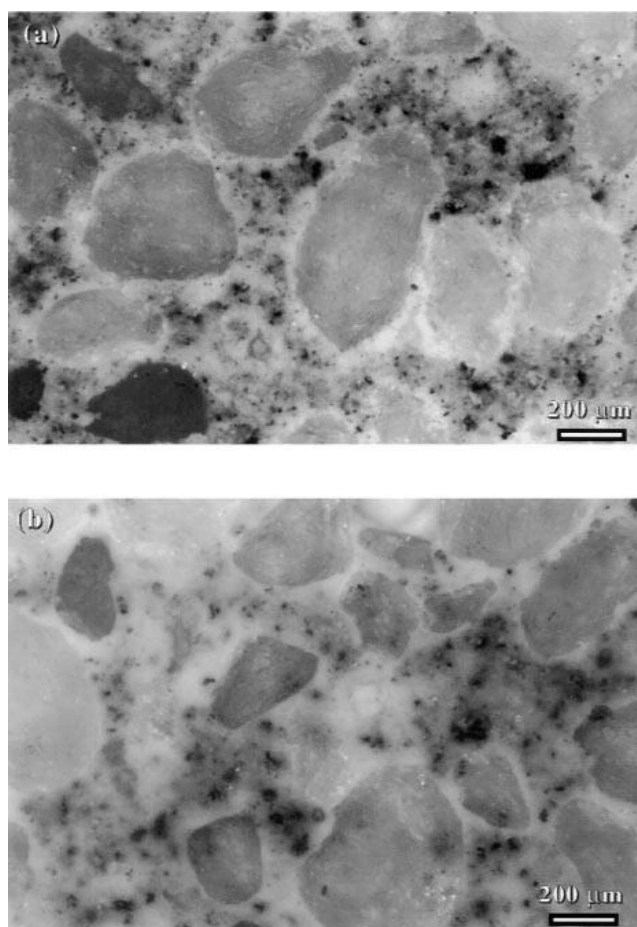
Fig. 1. Polarizing optical micrographs of the cut surfaces of mortars showing distributions of cement particles around sand grains. (*a*) Without PVA, (*b*) with 2 wt% PVA (based on cement). Hydration time, 28 days; w/ c ratio, 0.5.

greater PVA content. For example, the density had de-creased about 5% with 2 wt% PVA (based on cement mass). With 2.0 wt% PVA, the density was 2.04 mg/m^3; without PVA, the density was around 2.17 mg/m^3. The viscosity and the bleeding of mortar were both found to be reduced with the addition of PVA, as had been observed in PVA-modified cement pastes [7].

3.1.2. Mortar microstructure

The cut surfaces of the hardened mortar specimens were examined with both polarizing optical microscopy and scanning electron microscopy in backscattered mode. Typical micrographs are shown in Fig. 1 and Fig. 2. The cement particles are seen as small dark spots in the optical micro-graphs and as the brightest spots in the backscattered electron micrographs; the sand grains are the larger objects. In the optical micrograph without PVA (Fig. 1a), distinct white bands 30–80 mm wide are seen to surround each sand grain. Neighboring bands tend to overlap or connect be-
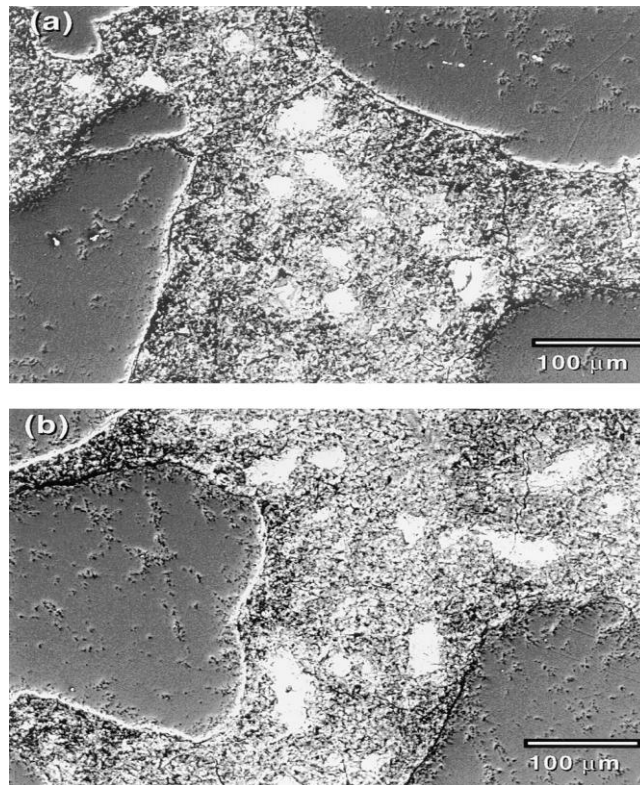
Fig. 2. Backscattered electron micrographs of the cut surfaces of mortars showing distributions of cement particles around sand grains. (*a*) Without PVA, (*b*) with 2 wt% PVA (based on cement). Hydration time, 28 days; w/ c ratio, 0.5.
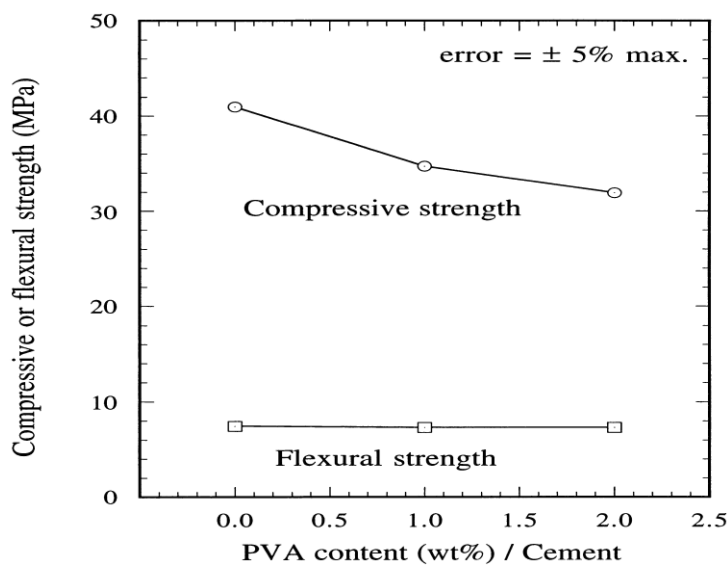

Fig. 3. Effect of PVA on the compressive and flexural strengths of mortar. Hydration time, 28 days; w/c ratio, 0.5.

cause of the width of the bands and the closeness of the sand grains. In the backscattered electron micrograph of the mortar without PVA (Fig. 2a), a degree of darkness is seen in the regions surrounding the sand grains within 30–80 mm of the grains. Because the backscattered brightness is proportional to material density, the darkness suggests higher porosity in these regions. In both the optical and backscattered electron micrographs of mortar without PVA, the cement particles that are visible tend to be distributed non-uniformly in the cement paste and to concentrate in the paste-rich regions between the sand grains and away from the sand grain surfaces. In the mortars with 2% PVA/cement (Figs. 1b and 2b), such bands surrounding the sand grains were less common, and when they did occur, they were thinner. As a result, neighboring bands tended not to overlap or connect but to be isolated. Also, the cement particles tended to be more uniformly distributed through the paste and were found adjacent to the sand grains as often as away from the grains. Macroscopically, PVA-modified mortar looked lighter in color and had more air voids, and both were observable with optical microscopy.

3.1.3. Mechanical strength tests

The compressive and flexural strengths of mortar with different amounts of PVA are shown in Fig. 3. The compressive strength of the mortar, measured with specimens 76 mm (3 in) in diameter and 152 mm (6 in) high, tended to decrease with PVA. The compressive strength decreased about 20% with 2% PVA/cement. The flexural strength of the mortar, measured by the four-point bending of square bars with the dimensions of 51 3 51 3 305 mm (2 3 2 3 12 in) and a span length of 229 mm (9 in), however, did not show any significant change with PVA content.

Concrete

Fresh concrete properties

Consistent with PVA-modified mortar, PVA-modified concrete had more air voids and lower density. Typical den-sity changes with the addition of PVA are shown in Figure. The density is seen to have decreased about 1% per 1 wt% PVA (based on cement mass).

IV DISCUSSION

The addition of PVA caused several changes in the micro structure and properties of mortar and concretes. The air void contents were increased, which decreased the mass density. The apparent viscosity and the bleeding of fresh mixtures were reduced. The porous interfacial transition zones around sand grains and coarse aggregate were reduced in size and number, and the unhydrated cement particles were distributed more uniformly in the cement paste phase, without significant depletion near aggregate surfaces in both mortars and concretes. For mortars, the compressive strength was decreased moderately, but the flexural strength was unchanged. For concretes, the compressive strengths after 28 days of hydration were relatively unchanged, even with their lower densities. However, the postpeak area un-der the compression stress-strain curve was reduced. The failure behavior had changed from a debonding of the coarse aggregate from the matrix to a cohesive fracture of the coarse aggregate. The compressive strength was modestly increased by PVA for a concrete having the same

den-sity as one without PVA. Some of the changes in behavior of mortar and concrete induced by PVA are related to those found for a model aggregate-paste bond system reported previously [6].

4.1. Interfacial transition zone in mortar and concrete

The reduction of the interfacial transition zone around sand grains and pea gravel in mortar and concrete with the addition of PVA (Figs. 1, 2) also seems to have arisen from the tendency for PVA to retard flocculation of the cement particles. Steric stabilization can also increase the efficiency of deflocculating by shear and vibration. The formation of a water-rich layer around aggregate surfaces is thus minimized or avoided [6].

Although not quantitatively measured, the hardened and dried PVA-modified mortar showed much slower absorption of water than did unmodified mortar when several droplets of water were placed on their cut surfaces. Water on the surface of PVA-modified mortar spread much less and stayed much longer. The reduction of the interfacial transition zone in the PVA-modified mortar seems to have contributed to the slower absorption of water, which is an indication of low permeability. The interconnected porous interfacial transition zones around sand grains in unmodified mortar are known to act as continuous channels, causing a high permeability of mortar and concrete [9]. By reducing the occurrence and thickness of the zones with PVA, the porosity becomes discontinuous, resulting in low permeability. The swelling of the PVA by water that has been used by Ohama to explain the low water permeability of methyl cellulose-modified mortar [5] may also have contributed to the lower permeability of the PVA-modified mortar. Because high permeability is known to reduce the durability of mortar and concrete [10], the addition of even small amounts of PVA to mortar and concrete, as in the present work, may improve the durability of these materials.

V CONCLUSIONS

The addition of small amounts of PVA (up to 2 wt% PVA based on cement mass) caused several changes in the microstructure and properties of mortar and concretes. The air void contents were increased, which decreased the mass density. The apparent viscosity and bleeding of fresh mixtures were reduced. The porous interfacial transition zones around sand grains and coarse aggregate were significantly reduced in size and number, and the unhydrated cement particles were distributed more uniformly in the cement paste phase, without significant depletion near aggregate surfaces in both mortars and concretes. For mortars, the compressive strength was decreased moderately, but the flexural strength was unchanged. For concretes, the compressive strengths after 28 days of hydration were relatively unchanged, even with their lower densities, but the postpeak area of the compression stress-strain curve was reduced. The fracture behavior had changed from debonding to cohesive failure of the coarse aggregate. The essentially similar strengths for mortar and concrete with and without PVA were obtained mainly because air void formation with PVA was minimized by using a prewetting mixing technique. When the same density with PVA as without was obtained, the compressive strength was moderately increased.

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