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Cement & Concrete Composites 21 (1999) 313–316

Cement & Concrete Composites

The interfacial zone and bond strength between aggregates and cement pastes incorporating high volumes of fly ash

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Received 6 January 1999; accepted 17 March 1999

Abstract

The weak transition zone between aggregate and cement paste controls many important properties of concrete. A number of studies dealing with interfacial zone are available in the literature for normal concrete and concrete containing silica fume. High-volume fly ash concrete for structural applications was developed at CANMET in the 1980s, but to date there has been no information available for interfacial zone in high-volume fly ash concrete.

In this paper, the orientation index and mean size of $Ca(OH)_2$ crystals in the aggregate-paste interfacial zone were determined by the X-ray diffractometer. The bond strength between the aggregate and paste was also investigated. It was found that, at the age of 28 days, there was no obvious transition zone between the aggregate and cement paste incorporating high volumes of fly ash. The higher the paste strength, the higher is the bond strength. © 1999 Elsevier Science Ltd. All rights reserved.

Keywords: Interfacial zone; Interface; Bond strength; Fly ash; Concrete; $Ca(OH)_2$ crystals; Orientation index; Mean size; High-volume fly ash; Paste; X-ray diffraction; Electron microcopy

1. Introduction

In 1956, Jacques Farran [1] was the first to observe a zone exhibiting a different mineralogy and microstructure at the aggregate-paste interface in concrete. In the last 15 or 20 yr, a number of researchers have demonstrated the existence of the transition zone. It has been found that the weak transition zone between the aggregate and paste controls many important properties of concrete [2], and the use of silica fume in concrete makes the zone dense and uniform [3].

High-volume fly ash (HVFA) concrete for structural applications was developed at CANMET [4]. Some investigations have also shown the potential use of the HVFA system for shotcreting [5] and lightweight concrete [6]. The investigation by CANMET and others have confirmed that HVFA concrete has many excellent properties [2,7]. However, the interfacial zone in HVFA concrete has not yet been studied. The purpose of this work was to investigate the interfacial zone and bond strength between the aggregates and the HVFA pastes.

2. Experimental

2.1. Materials

The cement used in all the mixes was portland cement. A Class I (Chinese Standard) fly ash from Chongqing, China, was selected for this work. The chemical analyses and physical properties of the cement and fly ash are presented in Table 1. The aggregate was limestone from Nanjing, China.

2.2. Paste mix proportions

Paste mixtures with different fly ash contents and water/binder ratios were prepared. The paste mix proportions used in this study are shown in Table 2. The consistency of each mix was standard consistency, except for mix F552.

2.3. Samples

The $10 \times 10 \times 20$ mm^3 aggregates were cut by a diamond saw. The surfaces of the aggregates were polished. One $10 \times 10 \times 20$ mm^3 aggregate was placed in each plastic mold. The pastes from each mix were cast in

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PII: S0958-9465(99)00013-X

Table 1
Chemical and physical properties of cement and fly ash

Chemical analysis (%)	Portland cement	Fly ash
SiO_2	21.42	44.98
Al_2O_3	4.68	30.08
Fe_2O_3	6.15	13.92
CaO	63.78	3.69
MgO	1.88	1.41
SO_3	1.08	0.15
Na_2O	0.19	–
K_2O	0.53	–
Loss on ignition	1.55	4.34
Specific gravity	3.16	2.38
Fineness >45 μm (%)	8.5	2.3
Compressive strength (MPa)		
3 days	36.8	–
7 days	45.9	–
28 days	59.2	–

Table 2
Paste mix proportions

Mix No.	*F*/(C + F) (%)	*W*/(C + F)	Admixture (%)
C0	0	0.26	–
F40	40	0.27	SA-1, 1.7; SA-2, 0.8
F551	55	0.28	SA-1, 1.7; SA-2, 0.8
F70	70	0.30	SA-1, 1.7; SA-2, 0.8
F552	55	0.38	SA-1, 1.7; SA-2, 0.8

these molds, respectively, and cured at 20 ± 3°C and more than 95% relative humidity. At the age of 28 days, the samples were demolded, and then separated at the aggregate-paste interface for X-ray diffraction analysis and scanning electron microscope analysis.

In addition, 40 × 40 × 160 mm^3 paste specimens were cast for compressive and flexural strength determination. In order to measure the bond strength between the aggregate and the paste, one 40 × 40 × 10 mm^3 aggregate cut with a diamond saw was put in the center of each 40 × 40 × 160 mm^3 mold, and the paste for each mix was then cast in the molds (Fig. 1). The samples were demolded at 1 or 2 days and cured in water at 20 ± 3°C until the time of testing.

2.4. Tests

1. X-ray diffraction (XRD) analysis of the solids: The samples were polished with emery paper layer by layer; XRD analysis of each layer was carried out using an Automated X-ray diffractometer (Cukα radiation, $2\theta = 10$–$70°$).
2. Scanning electron microscope (SEM) analysis: The morphology and microchemical features at the aggregate-paste interfacial zone were observed using a H800 scanning electron microscope with an energy dispersive X-ray analyzer (EDXA).

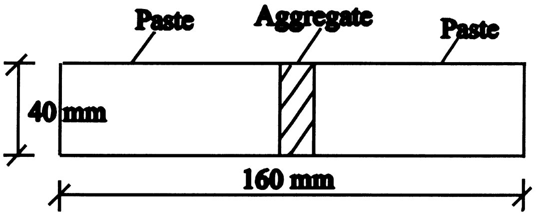

Fig. 1. Specimen for bond strength test.

3. Bond strength and paste strength: The bond strength between the aggregate and the paste, and the paste strength were measured according to GB177-85 (Chinese Standard). This standard is similar to European Standard EN 196-1:1987. The specimens are first tested in flexure so as to break into halves, then the broken halves are tested in compression as 40 mm equivalent cubes. The span of the flexural centre-point test is 100 mm.

3. Test results and discussion

3.1. Orientation index of $Ca(OH)_2$ crystals

It has been found that $Ca(OH)_2$ crystals tend to have their crystalline planes (0 0 1) parallel to the interface. This preferred orientation decreases away from the interface. In 1980, the degree of preferential orientation of $Ca(OH)_2$ crystals in aggregate-paste interfacial zone was measured by Grandet and Ollivier using XRD and a process of successive abrasion of the interface [8]. The orientation index of $Ca(OH)_2$ crystals defined by Grandet and Ollivier is:

$$R_{CH} = \frac{I_{(0\,0\,1)}/I_{(1\,0\,1)}}{0.74} \tag{1}$$

where $I_{(0\,0\,1)}$ and $I_{(1\,0\,1)}$ are the diffraction intensity values of crystalline planes (0 0 1) and (1 0 1) of $Ca(OH)_2$ crystals, respectively. If there is no preferential orientation, the ratio $I_{(0\,0\,1)}/I_{(1\,0\,1)}$ equals 0.74 and R_{CH} equals 1.0.

The relationships between orientation index and distance from the interface, for different mixtures at the age of 28 days, are shown in Fig. 2. It is seen that the paste made with portland cement only has a very high index of orientation. The orientation indices of the HVFA pastes are very low. There has been no obvious preferential orientation in the aggregates-HVFA pastes interfacial zone.

Test results [9] show that the content of $Ca(OH)_2$ produced by cement hydration in the HVFA paste is low. Some $Ca(OH)_2$ would also be consumed by the pozzolanic reaction. Therefore, the content of $Ca(OH)_2$ in the aggregate-HVFA paste interfacial zone is very low. In addition, the fine fly ash particles act as nuclei

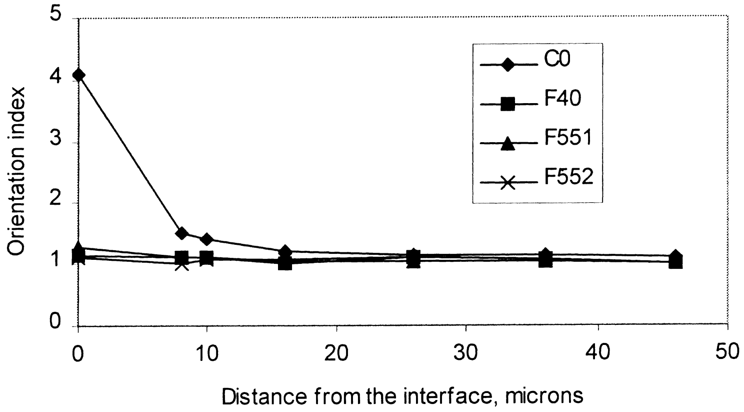

Fig. 2. Orientation index versus distance from the interface.

during the early stage of hydration and on which $Ca(OH)_2$ crystals grow. They would interfere with the growth and preferential orientation of $Ca(OH)_2$ crystals in the interfacial zone. These may be the reasons why there is no obvious preferential orientation in the aggregate-HVFA paste interfacial zone.

3.2. Mean size of $Ca(OH)_2$ crystals

Research results show that, in the aggregate-portland cement paste transition zone, the size of crystals is larger than that in bulk paste. In this study, the determination of mean size of $Ca(OH)_2$ crystals is based on the Scherrer equation [2]. The test results of the relationship between mean size of $Ca(OH)_2$ crystals normal to the crystalline plane (0 0 1) and distance from the interface are shown in Fig. 3. It is seen that, in the aggregate-portland cement paste interfacial zone, the mean size of $Ca(OH)_2$ crystals decreases with the distance from the interface. In the aggregate-HVFA paste interfacial zone, the mean size of the $Ca(OH)_2$ crystals is almost the same. These results imply that there is no obvious transition zone between aggregate and HVFA paste and there is an obvious transition zone between aggregate and portland cement paste. This is similar to the trend observed by the orientation index test.

3.3. SEM observations

Interfacial observations were made on the paste side for all the paste mixtures at 28 days. Test results reveal that there is an obvious preferential orientation of $Ca(OH)_2$ crystals for portland cement paste and there is no obvious preferential orientation for HVFA paste. There are also hydration product CSH and unhydrated fly ash particles at the aggregate-HVFA paste interface.

3.4. Bond strength

Results of bond strength (flexural strength) between the aggregate and the paste at different ages are given in Table 3. It can be seen from Table 3 that both the bond strength and the paste compressive strength increase with the age, and are affected by fly ash content and the water/binder ratio.

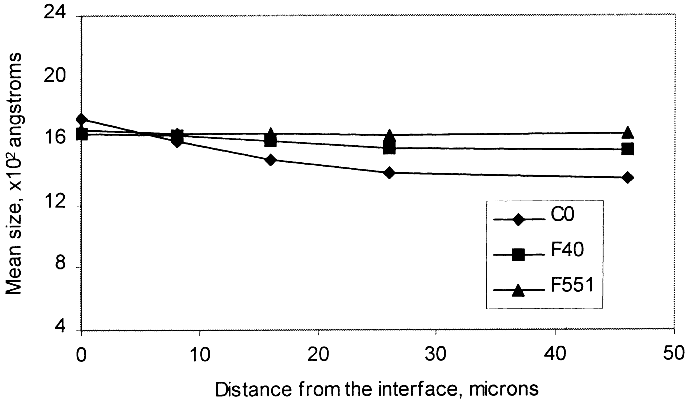

Fig. 3. Mean size versus distance from the interface.

Table 3
Bond strength between the aggregate and paste and compressive strength of pastes (MPa)

Mix No.	Bond strength			Paste compressive strength		
	3 days	28 days	90 days	3 days	28 days	90 days
F40	2.45	5.75	6.76	28.4	56.4	71.4
F551	1.63	3.59	5.41	20.0	37.1	64.1
F70	2.31	4.30	5.55	11.8	32.0	40.0

To analyze the bond strength in more detail, the ratio of the bond strength to the paste compressive strength is used. The results are shown in Table 4. The results of the ratio of the flexural strength to the compressive strength of the paste specimens (without aggregate) are shown in Table 5.

Comparing the results in Tables 4 and 5, it is seen that, at the age of 3 days, the ratios of bond strength to paste compressive strength for mixes F40 and F551 are lower than those of flexural strength to compressive strength, respectively. At the age of 28 and 90 days, both are almost the same. This implies that, at the age of 3 days, the interfacial zones for mixes F40 and F551 are weak. At the age of 28 and 90 days, there is no weak transition zone. For mix F70, there is no weak transition zone at 3, 28 and 90 days, but the absolute value of bond strength is still low at 3 days. Therefore, it can be concluded that there is no obvious weak transition zone between the aggregate and the HVFA paste after 28 days.

4. Conclusions

The following conclusion can be drawn from this study:

- There is no obvious preferential orientation of $Ca(OH)_2$ crystals in the aggregate-HVFA paste interfacial zone.
- The mean size of $Ca(OH)_2$ crystals is almost the same in the aggregate-HVFA paste interfacial zone.
- There is no obvious transition zone between the aggregate and the HVFA paste.
- At early ages, the interfacial zone between the aggregate and the HVFA paste is weak.
- The bond strength between the aggregate and the HVFA paste depends mainly on the paste strength. The higher the paste strength, the higher is the bond strength.

Table 4
Ratio of the bond strength to the paste compressive strength

Mix No.	3 days	28 days	90 days
F40	1/11.6	1/9.8	1/10.6
F551	1/12.3	1/10.3	1/11.8
F70	1/5.5	1/7.4	1/7.2

Table 5
Ratio of the flexural strength to compressive strength of the paste

Mix No.	3 days	28 days	90 days
F40	1/5.1	1/9.2	1/10.3
F551	1/5.5	1/10.4	1/10.4
F70	1/5.5	1/8.0	1/10.7

Acknowledgements

The author is grateful to Dr. V.M. Malhotra, CANMET, Ottawa, for his help in reviewing the final version.

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