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Influence of silane coupling agent on quality of interfacial transition zone between concrete substrate and repair materials

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Abstract

Silane coupling agent (SCA) was introduced as a modifying material to significantly improve the bond quality of the repaired interfacial transition zone. SCA aqueous solutions with various concentrations were used to coat the surfaces of a granite and of old concrete substrates before applying the repair materials. Both pull-off bond strength test and microstructure observation of the different repair interfacial layers were performed. The test results show that coating a concrete substrate with a SCA aqueous solution with an appropriate concentration can noticeably modify the microstructure of the interfacial transition zone, and therefore, significantly increase the bond strength.

Keywords: Silane coupling agent; Microstructure; Interfacial transition zone; Bond strength; Concrete substrate

1. Introduction

It has been reported that many applications of concrete repairs were not reliable though the necessary measures were taken to obtain as perfect adhesion as possible, such as roughening the old concrete and the application of a bond coat, including epoxy resin [1–3]. Silane coupling agent (SCA) is a kind of auxiliary for modifying the interfacial layers of composites. SCA molecules have multifunctional groups with a general chemical formula of $R\text{-}SiX_3$, where X stands for hydrolyzable groups bonded to Si, and R is a resin-compatible group. SCA is commonly used to significantly increase the bond strength and durability by providing the chemical bridge to connect the inorganic material (especially silicon-containing materials) and resin [4–6]. Can SCA provide a better bond between two inorganic materials (such as cement paste and stone)? An attempt to investigate this question was made by Ma [4]. He coated the surfaces of marble (with carbonate as the main constituent) specimens with styrene–butadiene resin emulsion, or KH-550, KH-560, KH-570 SCA solutions, separately, before applying cement mortar. Test results showed that the interfacial splitting tensile strengths of the modified interfacial layers were 27%, 57%, 69% and 84% higher than that of control specimens (using cement mortar only), respectively [4].

In view of this fact and considering that SCA can provide a better bond on a silicon-containing material surface [5–9], a research program to modify the repaired transition zone by using SCA solution to coat the surface of the concrete substrate before applying the repair materials was setup.

According to the experience of composite technology, concentration of SCA aqueous solutions has a significant influence on the bond strength of composites [7,9]. On the one hand, an aqueous SCA solution with a very low concentration may be not enough to create a SCA network that fully covers the surface of an inorganic material, resulting in a lower bond strength. On the other hand, an aqueous SCA solution with a very high concentration may induce a multiple molecular layer on the surface, creating a porous physically absorbent layer, leading to a

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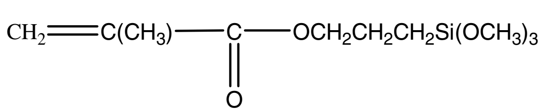
much lower bond strength [7,9]. Because no adequate information concerning the influence of the concentration of SCA aqueous solution on the bond strength between a concrete substrate and repair materials was available in current literature, an exploratory test to coat the concrete substrate by using 0.1%, 0.25% and 0.5% SCA aqueous solutions was carried out. The exploratory test results showed that 0.1% and 0.25% SCA solutions hardly affected the pull-off bond strength between the concrete substrate and the repair materials. SCA aqueous solutions with concentrations of 0.5%, 1% and 2%, therefore, were chosen for formal testing to evaluate the effects of the SCA aqueous solutions on the microstructure and bond strength of the repair interfaces.

2. Experimental program

2.1. Materials

Ordinary Portland cement (Chinese Standard GB175-1999, analogous with ASTM C 150) and a Class II fly ash (Chinese Standard GB15 96-91, analogous with ASTM C 618) were used in this research. The chemical analysis and physical properties of the cement and fly ash are presented in Table 1. Crushed stone with a maximum size of less than 20 mm and medium river sand with a fineness modulus of 2.44 were used for making concrete and repair mortar, respectively. Fine sand with a fineness modulus of 1.76 was used for producing primers. The mixture compositions of the concrete, repair mortar and primers are presented in Table 2. SCA, KH-570 with a molecular mass of 248.4, a relative density of 1.045, a boiling point of 255 °C and a refractive index of 1.429, from Nanjing, China, a commercially available coupling agent, was used. The molecular structure of KH-570 is presented by



2.2. Testing procedures

An ethyl alcohol aqueous solution with a mix ratio of 5:95 (ethyl alcohol:deionised water) by volume was first prepared. SCA aqueous solutions with concentrations of 0.5%, 1% and 2% were then made by using a magnetic stirrer. The mix ratios of these solutions were 0.5:99.5, 1:99 and 2:98 (SCA:ethyl alcohol/deionised water) by volume, respectively.

2.2.1. Preparation of substrate

Considering that the surface characteristics (such as porosity) of substrates greatly affect the effects of SCA solution [5–7], two old concrete beams 250 × 300 × 2500 mm aged six months and two granite planks 800 × 800 × 100 mm were used as substrates separately as shown in Fig. 1. As macro-roughness has a significant influence on bond strength and it is difficult to make the required parts (with or without using SCA) of the concrete beams having almost the same macro-roughness, the smooth surfaces of the concrete beams were prepared without any roughening operation in order to indicate the influence of SCA on bond properties better.

2.2.2. Application of repair mortars

A layer of each SCA aqueous solution was first brushed on the required parts of the surface of a concrete beam or granite plank as a substrate. After 12 h (to allow water evaporation), as shown in Fig. 1(a), 25 polyvinyl chloride (PVC) tubes with a diameter of 50 mm were placed on the surface of the concrete beam or granite plank as the formwork, a silicone glass adhesive was then applied at the outside of the PVC pipe bottoms to prevent the leakage of a repair material, i.e., a combination of a primer with or without the brushed SCA solution and a repair mortar. After another 12 h, a layer (about 3 mm thick) of M-primer (see Table 2) with or without the SCA solution, or F-primer was brushed in each tube on the surface of the concrete beam or granite plank, and the repair mortar was put in the brushed tube after not more than 30 min. This method

Table 1
Chemical and physical properties of cement and fly ash

Chemical analysis (%)	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	Na_2O	LOI	Blaine specific surface (m^2/kg)	28-day compressive strength (MPa)
Cement	19.9	4.3	6.12	65.7	1.54	1.05	0.32	1.41	465	46.7
Fly ash	52.8	18.9	8.1	14.5	1.78	0.31	0.46	3.58	573	–

Table 2
Mix proportions of concrete and repair materials (by mass ratios)

Type of material	Cement	Water	Crushed stone	Medium sand	Fine sand	Fly ash
Concrete	1	0.44	2.55	1.57	–	–
Repair mortar	1	0.44	–	2	–	–
M-primer	1	0.4	–		1	–
F-primer	1	0.44	–	–	1.1	0.1

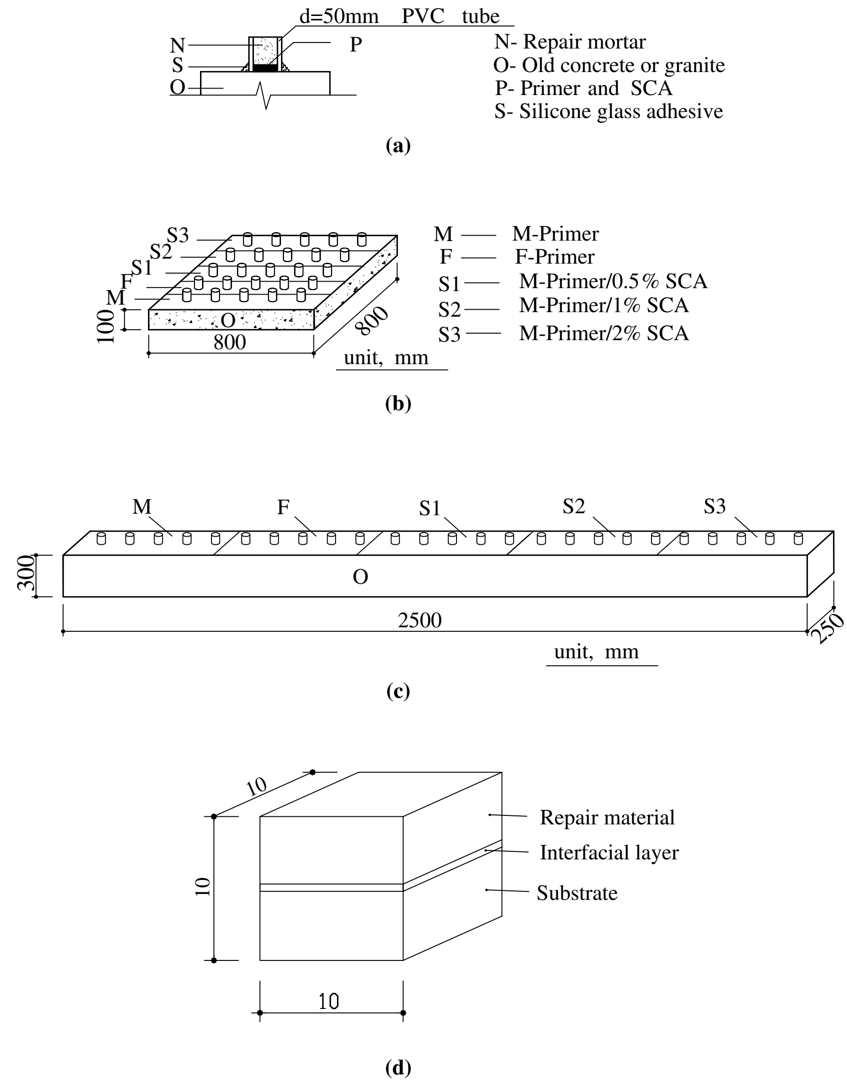


Fig. 1. Specimens for pull-off bond test and microstructure observation: (a) repair procedure; (b) granite plank specimen; (c) concrete beam specimen (d) sample for SEM observation.

does not need to drill core specimens from the repaired substrates, has no risk to damage the core specimens; and therefore is suitable for such comparison test. The repaired portions for all the specimens were wet-cured by covering with wet burlap for 28 days (Chinese Standard GBJ 81–85, analogous with ASTM C 192). After curing, every PVC tube was dismantled by using an electric iron to cut (melt) a narrow gap in the tube.

2.3. Pull-off bond test and microstructure observation

According to the different applied SCA solutions and primers, every old concrete beam and granite plank was divided into five parts as shown in Fig. 1 and Table 3. Each part had five specimens. One concrete beam and one granite plank were used for 28-day pull-off test for bond strength by using a DYNA pull-off tester. The other concrete beam and granite plank were used for scanning electron microscope (SEM) analysis. Three samples with a size of about $10 \times 10 \times 10$ mm^3 were taken from each specimen (see Fig. 1(d)). Every sample included a substrate, interfacial layer and repair material. Immediately after sampling, all samples were put into absolute alcohol, and gold-coated

Table 3
Influence of concentration of SCA on bond strength of repair interfaces

Interface type	Concentration of SCA (%)	Mean strength (MPa)	Coefficient of variation (%)
M-primer/granite	–	0.56	6.8
F-primer/granite	–	0.74	10.7
M-primer/0.5%SCA/granite	0.5	1.04	9.6
M-primer/1%SCA/granite	1.0	1.24	8.2
M-primer/2%SCA/granite	2.0	1.05	5.6
M-primer/concrete	–	1.13	12.1
F-primer/concrete	–	1.36	11.3
M-primer/0.5%SCA/concrete	0.5	1.56	12.9
M-primer/1%SCA/concrete	1.0	1.41	7.2
M-primer/2%SCA/concrete	2.0	0.75	10.2

before SEM observation. The interfacial transition zones were observed by a H-1030 SEM.

3. Results and discussion

3.1. Influence on bond strength

As shown in Table 3, the bond strength of M-primer/1%SCA/granite interface is 121.4%, 67.6%, 19.2% and 18.1% higher than that of M-primer/granite, F-primer/granite, M-primer/0.5%SCA/granite and M-primer/2%SCA/granite interfaces, respectively. This is the evidence that the use of SCA solutions results in a significant increase in the bond strength, and a 1% SCA solution creates a relatively better bond interface.

The bond strength of M-primer/0.5%SCA/concrete interface is 38.1%, 14.7%, 10.6% and 108% higher than that of M-primer/concrete, F-primer/concrete, M-primer/1%SCA/concrete and M-primer/2%SCA/concrete interfaces, respectively. The interface using a 2% SCA solution shows a sharp drop in the bond strength compared to that using a 0.5% SCA solution, probably because the higher concentration of the 2% SCA solution may induce a multiple molecular layer on the surface of the substrate, leading to a significant decrease in the bond strength [7,9].

The bond strength of the repair material/concrete interface is significantly higher than that of the repair material/granite interface. This is caused by the similarity of concrete and repair mortar, which promotes Van der Waals attraction forces.

The much higher increase of the bond strength on M-primer/SCA/granite interfaces compared to M-primer/SCA/concrete interfaces indicates that the SCA exhibits better effect on the granite substrate. The main reason for this may be because the relatively low-porosity granite substrate results in fewer SCA molecular layers to be created, and consequently leads to a higher increment of the bond strength [7,9]. The relatively high-porosity concrete substrate, however, may absorb more SCA solution and result in more SCA molecular layers.

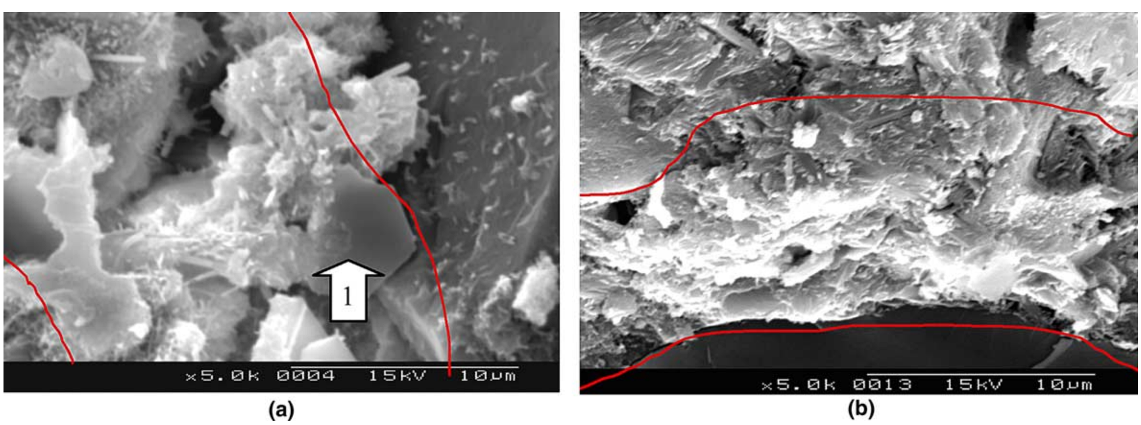


Fig. 2. SEM micrographs of M-primer/granite and M-primer/1%SCA/granite interfacial transition zones: (a) SEM micrograph of interfacial layer without using coupling agent solution; (b) SEM micrograph of interfacial layer by using 1% coupling agent solution.

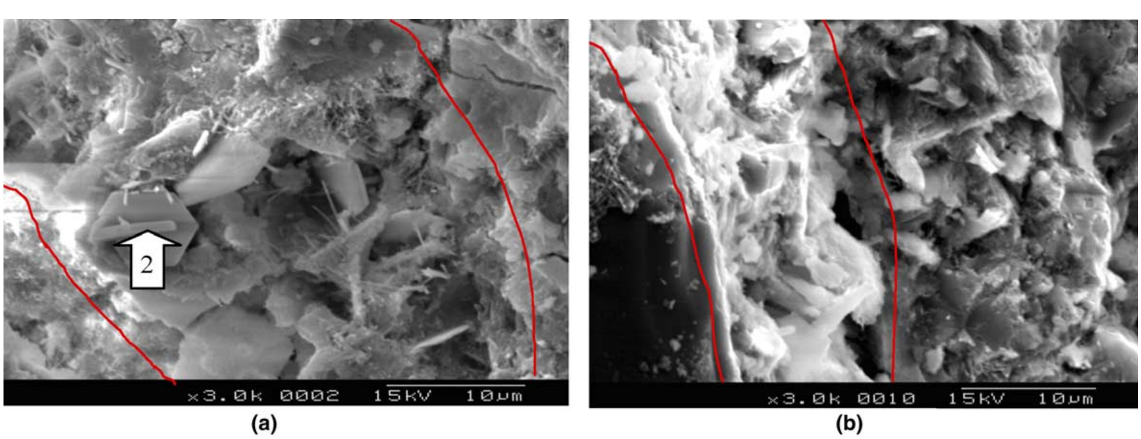


Fig. 3. SEM micrographs of M-primer/concrete and M-primer/0.5%SCA/concrete interfacial transition zones: (a) SEM micrograph of interfacial layer without using coupling agent solution; (b) SEM micrograph of interfacial layer by using 0.5% coupling agent solution.

3.2. Influence on microstructure of transition zone

3.2.1. Interfacial transition zone without using SCA

Some obvious separations between M-primer and granite or concrete substrate, induced by shrinkage could be observed by SEM as shown in Figs. 2(a) and 3(a). Both M-primer/granite and M-primer/concrete interfacial transition zones have about twice the thickness of SCA-modified ones. Both M-primer/granite and M-primer/concrete interfacial transition zones contain cement hydration products with larger crystals of $Ca(OH)_2$ (Arrows 1 and 2) and ettringite. The presence of the large crystals of $Ca(OH)_2$ indicates that the water content was high enough to allow large crystals being formed, leading to a higher porosity. No unhydrated cement particle could be observed, indicating that water/cement ratio at the interfacial transition zone is higher than that elsewhere. As a result, the bond strengths of the M-primer/substrate interfaces were much lower than those of M-primer/SCA/substrate interfaces (Table 3).

3.2.2. SCA-modified interfacial transition zone

The M-primer/1%SCA/granite and M-primer/0.5%SCA/concrete interfacial transition zones contain mainly C–S–H, and no clear crystalline morphology could be observed in Figs. 2(b) and 3(b), probably because the hydrophobicity of SCA network structure [8,9] makes large crystals hardly grow (due lower water content). As a result, SCA-modified interfaces contain far more C–S–H compared to the unmodified ones.

4. Conclusions

SCA can noticeably improve the microstructures of cement hydrates in the interfacial transition zone.

The use of SCA solution with a concentration in the range of 0.5–1% can improve the bond properties of repaired interfacial layer.

The modifying mechanism of the repaired interfacial transition zone by using SCA is worth further investigation.

Acknowledgement

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