Self healing mortars by using different cementitious materials

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ABSTRACT: This research investigates the self healing ability of mortars incorporating different cementitious materials. Ordinary Portland cement, crystalline additive (CA) and calcium-sulfoaluminate based expansive additive (CSA) were used in the study. The experimental results show the self crack closing phenomenon of all mortars, however, to different extent. The ability of crack closing was in the order of Control < CA1.5 < CSA10 < CA1.5/CSA10 where the numbers indicate the percentage of replacement. As observed from the external surface, the CA1.5/CSA10 which contained both CA and CSA shows a rapid self healing process which the surface crack up to 400 microns can be sealed within 28 days. The smaller surface cracks (<200 microns) can be sealed within 14 days. Water passing test confirmed the crack sealing ability of the mortars.

1 INTRODUCTION

Self-healing mechanisms in cement-based materials are currently based on further hydration of unreacted particles. These particles can be ordinary Portland cement as well as other types of conventional binding materials, which had not been fully hydrated in the initial stage of hydration, particular in low w/c mixes. There are various practical experiences and experimental studies (Wagner (1974), Ripphausen (1989), Edvardsen (1999)) which have demonstrated that the healing of crack in cementitious materials leads to a reduction of water permeability over time. However, such a mechanism is not highly promising in healing typical cracks in concrete because of the limited remaining potential and difficulties in optimizing the performance. Generally, the volume of hydration products of cement is not sufficient to close large cracks. The healing is limited to a crack width which is not larger than 50-150 μm. Li and Yang (2007) reported that the maximum crack width of 50 μm is necessary to achieve full recovery of mechanical and transport properties in engineered cementitious composite (ECC) material. Between 50-150 μm, partial recovery can be attained. In mechanical view, for cracked ECC specimens submerged in water, the deflection capacity recover about 65-105% from original specimens (Qian et al. (2009)).

Recently, various alternative cementitious materials are used in the research of self-healing concrete. Hosada et al. (2007), Kishi et al. (2007) and Sisomphon and Copuroglu (2010) investigated the self-healing potential of concrete when calcium sulfoaluminate based expansive agents (CSA) were used as a cement replacement material. Jaroenratanapirom and Sahamitmongkol (2010) evaluated the self-healing performance of mortars with different additives namely, fly ash (FA), silica fume (SF) and crystalline admixture (CA). Qian et al.
(2010) reported the self-healing ability of engineered cementitious composite (ECC) incorporating limestone powder, blast furnace slag and nanoclay.

In this study, the self-healing potential of mortars incorporating the combined use of CSA and CA was evaluated. The effect of additives on microstructure and permeability of cracked mortars was investigated. Quantitative analysis on residual surface crack width was chosen as a method for determining of damage and healing degrees.

2 EXPERIMENTS

Portland cement CEM I 42.5 N, crystalline admixture and expansive additive were used in this study. Crystalline admixture (CA) is a synthetic cementitious material which contains reactive silica and some crystalline catalysts. Some chemical components react with Ca(OH)$_2$ to form crystalline products which disconnect pores and fill cracks in the concrete. The crystalline products can only occur when sufficient moisture is present. The main application of crystalline material is to improve the water tightness and to stop leakage of concrete structures. It is also classified as one of the hydrophilic water-proofing materials (Yodmalai et al. (2010)). Apart from CA, a synthesized ternary blend (e.g. Hauyne, anhydrite and free lime) calcium sulfo-aluminate based expansive additive (CSA) which is a commercial product normally for shrinkage compensation was used.

All mixtures were designed based on water-to-cementitious ratio ($w/cm$) of 0.25 and sand-to-cementitious ratio ($s/cm$) of 2.0 by mass. The mix designs of mortars are demonstrated in Table 1. Superplasticizer, Glenium ACE30, was used to achieve workable condition of mortars. To prepare specimens, the mixing sequence was 2 min low speed followed by 2 min high speed mixing with a commercial Hobart mixer. The disc-shape specimens were cast in plastic containers with 75 mm diameter and a height of about 20 mm. To achieve large crack width, all specimens have to be reinforced with galvanized wire-mesh which was place at the mid-height of the specimens. The filled containers were vibrated for 20 s on a vibrating table. After 24 h, specimens were demolded and subsequently damp cured at a controlled temperature of 25 ± 2°C and 95 ± 5% RH for a 3 day. Eventually, all specimens were exposed to laboratory air for another 25-day period.

<table>
<thead>
<tr>
<th>Mix</th>
<th>OPC</th>
<th>CSA</th>
<th>CA</th>
<th>Water</th>
<th>Sand</th>
<th>Superplasticizer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>100</td>
<td>-</td>
<td>-</td>
<td>25</td>
<td>200</td>
<td>2</td>
</tr>
<tr>
<td>CA1.5</td>
<td>98.5</td>
<td>-</td>
<td>1.5</td>
<td>25</td>
<td>200</td>
<td>2</td>
</tr>
<tr>
<td>CSA10</td>
<td>90</td>
<td>10</td>
<td>-</td>
<td>25</td>
<td>200</td>
<td>2</td>
</tr>
<tr>
<td>CA1.5/CSA10</td>
<td>88.5</td>
<td>10</td>
<td>1.5</td>
<td>25</td>
<td>200</td>
<td>2</td>
</tr>
</tbody>
</table>

The specimens were pre-cracked in the manner of splitting tensile strength test to crack width of about 100-400 microns (Fig. 1). After pre-cracking, the specimens were fastened as tightly as possible by a 75mm diameter PVC pipe bracket to create a confinement. The purpose of this process was to imitate a real condition where hardened concrete is usually under some constraint in the real structure. Stereomicroscope with computer software was used to evaluate surface crack widths. The crack width was measured from 5 positions on each surface of specimens. For each mix, two pieces of disc-specimens were used for analysis. Hence, there were totally 20 positions of crack which were measured and monitored for each mixture. Distance between each position was about 10 mm as showed in Fig. 2. Initial crack widths were measured immediately after bracket installation. Then, all specimens were submerged into water which was replaced once every 7 days. The residual surface crack widths were measured at 4, 7, 14 and 28 days wetting periods. As water was replaced every 7 days, it has to be noted that the crack width measurement was performed at the stage before water renewal.
Water passing test was conducted on pre-cracked mortars to investigate the permeability of mortar due to crack progression. The specimen was attached to a plastic cylinder, and tightly fastened with a PVC bracket. Silicone is applied along the seam to prevent water leakage (Fig. 3a). A constant water head of 100±5 mm was controlled throughout the experiment (Fig. 3b). The specimen was placed on supports to allow free movement of water. The water passing rates were measured at 4, 7, 8, 14, 15 and 28 days wetting periods. The duration of each measurement was 10 minutes.

Eventually, the appearance of healed cracks was observed internally. The profile of crack and formation of healing products were studied on polished specimens by using a stereo microscope. The integrity of the samples was maintained by impregnation with a low viscosity fluorescent dye epoxy under vacuum. The specimens were vertically sliced perpendicular to the crack direction to a thickness of about 10 mm by a machine saw and dried in the oven at 35°C for 24 h. Thereafter, the sliced specimens were impregnated in dyed epoxy under vacuum for another round. After the hardening of epoxy, the specimens were ground by using DBT Diamond Roller and Grinder/86 thin sectioning unit for initial preparation of polished sections. Finally, specimens were polished with 6 μm, 3 μm, 1 μm and 0.25 μm diamond pastes, respectively.

3 RESULTS AND DISCUSSION

The surface cracks of mortars before and after self healing process were investigated by stereo optical microscope at 40× magnification. Fig. 4 shows the development of crack closing process with respect to wetting period. The width of crack was measured on 20 different pre-specified locations at 0, 3, 7, 14 and 28 days wetting period. The initial crack width of all specimens was found in a range between about 50 and 400 microns. Generally, there was no visible difference of crack sealing rates between those on top surfaces and bottom surfaces for each particular mix. The change of surface crack width was used as an indicator of quantitative evaluation of self healing degree. The reduction of crack width of all specimens is shown in Fig. 5(a-d).
Figure 4. Investigation of crack closing with respect to wetting period.

Figure 5. Crack width reduction of control mix corresponding to wetting period (initial crack width: >300\,\mu m (bold solid lines); 200-300\,\mu m (dashed lines); <200\,\mu m (thin solid lines)).

As demonstrated in Fig. 5, the cracks were categorized into 3 groups of crack width represented by bold solid lines, dashed lines and thin solid lines which were for the initial crack width of >300 \, \mu m, 200-300 \, \mu m and <200 \, \mu m, respectively. The results indicate that the cracks of all mixtures were healed with different rates. The ability of crack closing was in the order of Control < CA1.5 \approx CSA10 < CA1.5/CSA10. The self healing ability can be observed on control mix, while the crack up to about 150 \, \mu m width has been closed within 28 days. However, the mortars with CSA and CA showed superior self-healing ability. Generally, there
was no significant difference between the healing abilities of CA1.5 and CSA10 mixes. It is clearly seen that the CA1.5/CSA10 mixture showed the optimum healing potential where the surface crack up to about 400 microns can be sealed within 28 days.

The average of relative crack width for different ranges of initial crack width was demonstrated in Fig. 6(a-c). For small initial cracks (<200μm), the residual crack width of the control mortar was about 10% of the initial width after 28 days wetting. It can be seen that CA1.5, CSA10 and CA1.5/CSA10 mortars can close the small cracks within 14 days. However, only CA1.5/CSA10 mix has a potential to completely seal medium (200-300μm) and large cracks (>300μm) within about 28 days. In this view, it can be obviously seen that CSA10 mix has a higher healing potential than the CA1.5 mix at the first 14 days. At 28 days wetting, however, there was almost no difference between CA1.5 and CSA10 mixes. However, a more rapid healing ability would be more preferable in practice. Hence, the ability of surface crack closing would be ranked in the order of Control < CA1.5 < CSA10 < CA1.5/CSA10.

![Figure 6. Effect of initial crack width on the crack width reduction.](image)

Fig. 7 shows the result of water passing rate of pre-cracked mortars corresponding to wetting period. At the beginning of the test, the initial water passing rates were measured. Thereafter, the reduction of water passing rate was presented in relative values to the initial water passing rate. For the control mortar, slight reduction of water passing rate can be observed in the early period, while those mortars with CA or CSA showed rapid reduction of water passing ability at the first 5 days. The rates of reduction were gradually decreased between 5 and 7 days. However, after the water renewal at 7 days, the water passing rates of all mortars obviously rebounded. From this observation, it can be hypothesized that some healing products can be dissolved or decomposed under fresh water exposure. While most concretes in practice are usually exposed to fresh water, it is important to note that the specimens continuously released Ca^{2+} and OH^{-} to the curing water. Owing to the restricted environment of curing tank in laboratory, the rather high alkalinity of curing water in laboratory experiment may cause the misleading evaluation of concrete self-healing phenomenon, particularly the surface crack closing ability. The water in curing tank would be frequently replaced. Ideally, the pH of curing water should be kept at about neutral throughout the wetting phase.
Figure 7. Relative water passing rate of pre-cracked mortars.

The microstructures of cracks and healing products in cross-sectional view were observed on polished specimens. At the near surface area, it can be seen from photomicrographs that the major product was characteristic of calcium carbonate (Fig. 8a). The appearance of crack closing seen on the surface (Fig. 4) would be majority due to the precipitation of calcium carbonate which deposited on the surface of specimens. In previous work (Sisomphon and Copuroglu (2010)), the microstructures of crack healing minerals were observed on petrographic thin-section through a polarized light microscope, and it was also clearly revealed the formation of calcium carbonate on surface crack (Fig. 8b).

In a previous study, similar experiments were conducted on disc-mortars without any confinement. It was noticed that there are numerous micro-cracks in the specimens, particularly at the interfacial transition zones (ITZ) between matrix and aggregate (Fig. 9a). Generally, the size of crack width was about 20-30 μm. This would be due to the effective expansion of the matrix with CSA addition. Microcracks would appear, if the expansive strain of the matrix exceeds its tensile strain capacity. Because ITZ is usually weaker than the bulk paste, microcracks can be found around sand particles. In this study, however, the specimens were fastened by pipe brackets to create a confinement. The purpose of this process was to imitate a real condition where hardened concrete is usually under some constraint in the real structure. The confinement would improve self healing ability. Moreover, the specimen could not be freely expanded; hence, microcracks on ITZ can be minimized (Fig. 9b). Although the main crack in Fig. 9a seems to be smaller than that in Fig. 9b, it has to be noted that the prior had a smaller initial crack width than the latter.

(a) Polished specimen under UV light (b) Thin-section (Sisomphon and Copuroglu (2010))

Figure 8. Precipitation of calcium carbonate on surface crack.
For internal cracks, it was observed that the crack widths of the mortars with CSA were decreased. The expansion of the bulk matrix due to ettringite formation would play a role on the contraction of crack width. Ettringite formation on crack wall and also in the bulk matrix can be seen (Fig. 10). For external appearances, the experimental results indicated that the surface crack closing phenomenon would be mainly due to the formation of calcium carbonate. However, it has to be noted that a significant more amount of calcium carbonate can be observed on the mortars with CSA addition. Therefore, it is possible that the carbonate products could not be calcium carbonate alone. This could be involved with the reaction of ettringite with carbon dioxide which yield carbonate analog of ettringite. As another hypothesis, the net-like formation of ettringite on crack walls would serve as a support, and allow more precipitation of calcium carbonate. These hypotheses have to be clarified in future studies.

4 CONCLUSIONS

This paper presents the results on the crack closing potential of mortars incorporating calcium sulfoaluminate based expansive additive (CSA) and crystalline additive (CA). The following conclusion can be given based on the laboratory experiments in this study:

- The usage of CSA and CA is beneficial with respect to crack closing phenomenon. The combined use of 10% CSA and 1.5% CA replacement revealed the optimized healing potential.
• The experimental results indicated that CA1.5, CSA10 and CA1.5/CSA10 mortars can close the small cracks (<200μm) within 14 days. However, only CA1.5/CSA10 mix has a potential to completely seal 200-400μm cracks within about 28 days.

• It seems that the physical of net-like formation of ettringite is beneficial to the formation of calcium carbonate, particularly at the near surface area.

• After 28 days wetting, the water passing rates of pre-cracked mortars with CSA and CA have decreased to <10% of the initial values.

• Confinement is necessary for the self healing experiment of mixtures with expansive additive. The confinement would improve self healing ability, and minimize microcrack on the interfacial transition zone between matrix and aggregate. The confinement was also to imitate a real condition where hardened concrete is usually under some constraint in the real structure

ACKNOWLEDGEMENTS

The authors would like to thank Agentschap NL for the financial support granted for the project SHM08729, as well as materials and technical supports from Denka Chemical GmbH and Xypex Corporation.

REFERENCES


