Construction and Building Materials 189 (2018) 1054–1066

Contents lists available at ScienceDirect

Construction and Building Materials

journal homepage: www.elsevier.com/locate/conbuildmat

Self-healing capability of cementitious materials with crystalline admixtures and super absorbent polymers (SAPs)

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HIGHLIGHTS

- The self-healing property of cementitious materials was investigated.
- Different crystalline admixtures and super absorbent polymers were considered.
- Water flow and crack closing tests were performed followed by SEM image analysis.
- CAs increased the self-healing capability, while SAPs swelled to seal cracks.
- Using CAs with SAPs accelerated self-healing.

ARTICLE INFO

Article history:
Received 9 November 2017
Received in revised form 1 August 2018
Accepted 12 September 2018

Keywords:
Self-healing
Water flow test
Cementitious material
Crystalline admixture
SAPs

ABSTRACT

In this study, the self-healing property of cementitious materials with crystalline admixtures (CAs) and super absorbent polymers (SAPs) was investigated. Specimens were pre-cracked with a crack width of 0.210–0.304 mm at 7 and 28 days. The self-healing capacity was evaluated with the water flow test and crack closing test, and self-healing products were investigated with a scanning electron microscope (SEM). The water flow of specimens containing CAs greatly decreased relative to that of the plain specimen after self-healing. The results indicate that the CAs increased the self-healing capability of cement-based materials. The specimen containing SAPs was observed to re-swell and seal cracks. For example, the water flow was reduced 100% in the case with a 0.256 mm crack width. Using CAs with SAPs accelerated the crack sealing. SEM observation confirmed that CAs produced more self-healing products around SAPs.

1. Introduction

In concrete structures, self-healing refers to the property of the structure being able to autonomously and autogenously repair minor damages or cracks caused by external loads and environmental factors. Many studies have recently been conducted on self-healing concrete so that structures can autonomously heal cracks to increase their service lives [1–3].

Previous studies have shown that cement-based materials can self-heal fine cracks. This is called autogenous healing because the self-healing property is intrinsic to the cement [4–6]. Autogenous healing is divided into two types of mechanisms. The first is crack filling caused by the further hydration of unhydrated cement. This occurs because water penetrates the concrete after a crack occurs and hydrates unhydrated cement-based materials on the crack surface. The additional reaction products formed by this process fill the crack space [7]. The second type is the precipitation of calcium carbonate crystallized by carbonation in a crack [7].

Autogenous healing can only fix very small crack widths. Various studies have been conducted on improving the self-healing of cementitious materials according to the binder type and mixing conditions to increase the durability of structures [8–15]. Granger et al. [8] confirmed that a lower water/binder (W/B) ratio produced more unhydrated material in the cement matrix, which increases the self-healing capability. Tittelboom et al. [9] used isothermal calorimetry to analyze the self-healing potential according to the mixing amounts of ordinary Portland cement (OPC), ground granulated blast-furnace slag (GGBS), and fly ash (FA). They confirmed that mixing in GGBS increased the self-healing potential, but mixing in FA reduced it. On the other hand, Liu et al. [10] reported that mixing FA in concrete improved the self-healing performance. This is similar to the results of Sahmaran et al. [11], who performed an experiment on the compressive strength recovery rate of cracked

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https://doi.org/10.1016/j.conbuildmat.2018.09.061
0950-0618/

self-consolidating concrete and showed that the self-healing capability improves with the amount of FA in the mixture. Qian et al. [12] investigated the deflection recovery rate of cracked beams through the four-point bending test and confirmed that GGBS and limestone powder improve the self-healing capability. Liu and Zuo [13] reported that the optimum self-healing capability was achieved for a mixture of 30% GGBS and 40% FA. Roig-Flores et al. [14] experimentally evaluated the self-healing capability with crystalline admixtures (CAs) under various environmental conditions; they confirmed that moisture is required for self-healing and that mixing in CAs improves the self-healing capability. Sisomphon et al. [15] performed a crack-closing test and reported the self-healing potential of cementitious materials mixed with a calcium sulfoaluminate (CSA)-based expansion additive and CAs.

As a similar concept to self-healing, self-sealing has been actively studied to prevent leakage in structures [16–20]. When moisture penetrates a structure after a crack occurs, the crack is sealed by the expansion of the swelling material on the crack surface. Superabsorbent polymers (SAPs) are the most widely used material for self-sealing, and many researchers have studied self-healing concrete using SAPs [17–22]. Mignon et al. [17] conducted a study on pH-responsive SAPs whose swelling properties vary with the pH in concrete pores. Lee et al. [18] performed the water seepage test to evaluate the self-sealing performance of SAPs and found that cracks larger than 0.3 mm could be healed. SAPs not only affect the self-sealing effect of cracks when mixed with concrete but also induce internal curing by absorbing moisture and releasing the absorbed moisture during the concrete curing [19]. As a result, mixing in SAPs should improve the self-healing performance of unreacted inorganic binders. Tittelboom et al. reported that water adsorbed by SAPs promotes further hydration of cement-based materials as it is slowly released during hydration [20].

When the crack width is large (0.3 mm or more), healing a crack only through self-healing by the further hydration of inorganic binders and precipitation of calcium carbonate is difficult. When the self-healing performances of SAPs and inorganic binders are applied together, however, it is expected that larger cracks can be healed than when only one type of material is used. In this study, the self-healing properties of cement composites were investigated according to the use of supplementary cementing materials (SCMs), a CSA expansion agent, CAs, and SAPs. For the experiment, paste specimens mixed at a W/B ratio of 0.3 were prepared, and 0.210–0.304 mm cracks were induced after 7 and 28 days. The self-healing capability was evaluated with the water flow and crack closing tests, and the self-healing products were analyzed with a scanning electron microscope (SEM).

2. Experiment

2.1. Materials

In this study, SCMs such as GGBS and FA were used as inorganic binders with OPC. A CSA expansion agent; $CaSO_4$, Na_2SO_4, and $MgCO_3$ ($MgCO_3{\cdot}Mg(OH)_2{\cdot}5H_2O$); and the commercial product Xypex were used as CAs. Fig. 1 shows the X-ray diffraction (XRD) analysis results for OPC, GGBS, FA and CSA. In the case of OPC, the main constituents were gypsum, calcite, and quartz in addition to C_3S, C_2S, C_3A, and C_4AF. Because GGBS is mostly composed of amorphous materials, only anhydrite and quartz were observed in the XRD pattern analysis.

A was found to mostly consist of quartz and mullite. This is because SiO_2 and Al_2O_3 are the main components of FA. In the XRD patterns of CSA, gypsum was the most dominant, and CaO, C_3S, and others were evenly distributed. In the XRD patterns of xypex, the main constituents were brucite, gypsum, calcite, and quartz in addition to C_3S, C_2S, C_3A, and C_4AF similar with OPC. However, the peak of Calcite and quartz was higher than that of OPC.

Tables 1 and 2 present the chemical compositions of the raw materials according to X-ray fluorescence (XRF) analysis and the physical properties of inorganic binders.

Fig. 2 represents the particle size distribution of the raw materials. The mean particle sizes of OPC, GGBS, FA, CSA and XYPEX were 19.1, 15.0, 26.9, 21.9 and 33.0 μm, respectively, and that of OPC ranged from 2.98 μm to 88.5 μm. The particle size of GGBS ranged from 1.98 μm to 59.9 μm, and that of FA showed a wide distribution from 2.98 μm to 152 μm. The particle size distribution of CSA was the widest and ranged from approximately 1 μm to 150 μm. The particle size distribution of XYPEX was the widest and ranged from approximately 5 μm to 262 μm.

In this study, SAPs produced by LG Chem Ltd. in South Korea through the bulk gel polymerization method were used. The SAP powders mainly consisted of acrylic acid, and their particle shapes were irregular because they were crushed in the final production stage. As shown in Fig. 3, three types of SAP powders were used according to the crushed particle size. The mean particle sizes and standard deviation of SAP were 541 μm and 57 μm for SH-SO1 and SH-SA1, 234 μm and 24 μm for SH-SO2 and SH-SA2, and 83 μm and 8 μm for SH-SO3 and SH-SA3 respectively, in the dried condition. Fig. 4 shows the SEM image of the SAP powders, which confirms that the particles were shaped irregularly like broken glass. A typical method for evaluating the absorption properties of SAPs is the teabag method and the filtration method. The teabag method is more effective to evaluate time-dependent absorption characteristics by 24 h. The filtration method is effective for predicting absorption characteristics after 24 h of immersion [23,24]. In this study, the teabag method was used to evaluate the absorption properties of SAP powders, and they could absorb approximately 250 mL of distilled water per 1 g of SAPs.

Table 3 presents the mixture proportions of the paste specimens. The W/B ratio was fixed at 0.3. SAPs corresponding to 0.5% of the binder weight were mixed in based on the results of the existing literature analysis. [25] For the specimens mixed with SAPs, extra water was added to consider internal curing due to SAP mixing. In this study, extra water corresponding to a W/B ratio of 0.05 was added based on the results of Shen et al. [26]. The SAPs were placed in a binder, mixed for about 2 min with a pan mixer, and mixed again for about 3 min after water was added.

2.2. Specimen preparation and test method

In this study, 50 mm × 25 mm disk specimens were prepared, cured in a moist-curing chamber at 20 ± 3 °C and RH 100% for 24 h after mixing, and then cured in a water bath maintained at 20 ± 3 °C until cracking. Cracks were created with the crack inducing method shown in Fig. 5(a). The specimens were completely separated into two pieces, and 0.2 mm diameter copper wires were inserted into the crack surface, as shown in Fig. 5(b), to maintain the crack width. Then, the separated pieces were reassembled. The peripheral parts of the specimens were then sealed with epoxy to fix the crack width. To prevent variations in the crack width during the experiment, the specimens were placed in a transparent acrylic pipe, and the space between them was fixed with an epoxy resin. The water flow test was performed after the epoxy resin completely hardened.

The crack widths of the specimens were measured with an optical microscope. Table 4 presents the mean and standard deviation values for the initial crack widths of the measured specimens. In the case of the specimens without SAPs, crack widths ranging from