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Rapid self-sealing of cracks in cementitious materials incorporating superabsorbent polymers

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HIGHLIGHTS

- Rapid self-sealing of cracks by SAPs in cementitious materials was investigated.
- Rapid swelling of SAPs effectively sealed the cracks within five minutes.
- Most SAPs were split in the cracks while remaining bonded to the surrounding matrix.
- Intact SAPs swelled across voids formed by swelling of SAPs including the cracks.
- Split SAPs swelled only as much as the volume of voids by SAPs not including crack.

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ABSTRACT

The effect of superabsorbent polymers (SAPs) on rapid self-sealing of cracks in cementitious materials was investigated experimentally. Rapid swelling of SAPs effectively sealed cracks in materials within five minutes, resulting in the reduction ratio of water runoff per unit time in ranges of 34–52% and 52–72% for SAP dosages of 0.5% and 1.0%, respectively. X-ray computed tomography (CT) analysis showed that the swelling ratio of SAPs in the specimens was less than that in the filtered cement pore and synthetic solutions. Analyses of images obtained using X-ray CT, cryofracture scanning electron microscopy, and optical microscopy indicated that when a crack occurred, most SAPs, which were split because of the crack while remaining bonded to the surrounding cement matrix, swelled only as much as the volume of voids formed by swelling of SAPs, not including the crack. However, a part of the SAPs that remained intact swelled across voids, including the crack.

1. Introduction

In recent years, extensive research has been performed on concrete that self-heals cracks [1]. With the aim of enhancing engineered self-healing of cementitious materials, several approaches, which are primarily microcapsule-based [2], bacteria-based [3], and mineral admixture-based, are being developed. In mineral admixture-based approaches, self-healing of cementitious materials is enhanced by effects of mineral admixtures such as swelling, expansion, and re-crystallization [4]. An existing study [5] has demonstrated that cracks with widths of up to 0.3 mm can be healed using the mineral admixture-based method; however, considerable time is required to heal cracks. Therefore, to improve healing efficiency in materials, superabsorbent polymers (SAPs) are used as an additional admixture with existing approaches [6,7].

While SAPs have been primarily used as internal curing agents to mitigate autogenous shrinkage of high strength cementitious materials [8–10], existing studies [11–13] suggest that SAPs can effectively seal cracks in materials when ingress water is provided through the cracks. SAPs are hydrophilic polymers that can swell significantly by absorbing and retaining a large amount of liquid [14–16]. SAPs swell in cementitious materials shortly after mixing by absorbing the water in fresh cementitious mixtures. However, this swelling is less than that caused by absorbing fresh water [17]. This is because ions, such as Ca^{2+}, K^{+}, Na^{+}, and OH^{-}, dissolve in water quickly during mixing [18], resulting in higher ionic concentration in mixing water than in fresh water and high pH (12.5–13.0), thereby impeding SAPs' ability to swell [19,20]. In addition, the decrease in the swelling capacity of SAPs results from the charge screening effect of the cations (K^{+}, Na^{+}, Mg^{2+}, and Ca^{2+}) and complex formation between divalent/trivalent ions (Ca^{2+} and

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Mg^{2+}) and carboxylate groups [13]. Over time, cementitious materials become dry and SAPs shrink by releasing absorbed water, producing voids in the materials [21]. As cementitious materials are likely to crack along these voids, SAPs inside the materials will be exposed to crack surfaces. When water penetrates the cracks, the dry SAPs in the cracks is expected to absorb this water and swell again [21]. As the ionic concentration of the ingress water provided to the cracks is typically lower than that of the water in fresh mixtures [22,23], swelling of SAPs will be considerably higher over crack surface, including the voids formed by SAPs during initial mixing, resulting in self-sealing of cracks in cementitious materials [21].

In recent years, several studies [11,21,24–28] have been conducted to develop methods of using SAPs to enhance the efficiency of self-sealing of cracks, i.e., sealing of cracks by swelling of SAPs, and self-healing of cracks, i.e., healing of cracks by formation of healing products, which is promoted by SAPs along the cracks, in cementitious materials. Lee et al. [11] found that a specimen incorporating 5% SAP by weight of binder reduced cumulative flow from cracks by 80% or more, as compared to a reference specimen without SAPs. Additionally, they proposed a self-sealing mechanism of cementitious materials incorporating SAPs [21]. Snoeck et al. [13,24] investigated self-sealing of SAP-added cementitious materials through neutral radiography and low-pressure water-permeability tests. Snoeck et al. [24] and Wang et al. [25] investigated enhancement in self-healing efficiency of cracks in cementitious materials, including different dosages of SAPs mixed with microfibers and bacteria. They found that $CaCO_3$ formed in the cracks and healed them. In addition, Snoeck et al. [26] analyzed 3D X-ray computed tomography (CT) images to determine the manner in which healing products such as $CaCO_3$, through which cracks were healed, were primarily formed in the cracks by SAPs under wet/dry cyclic conditions. Wang et al. [27] investigated the distribution of healing products in the cementitious materials that used hydrogel encapsulated bacterial spores by employing 3D X-ray CT. Lee et al. [28] proposed a model to determine the volume fraction of cracks sealed by SAPs in cementitious materials, in which parameters such as SAP dosage, swelling ratio, and crack width were included. Even though several studies [11–13,21,24–28] have been carried out to investigate the efficiency of self-sealing and self-healing of SAP-added cementitious materials, rapid self-sealing of cracks, which results from rapid swelling of SAPs by ingress water provided for the first time through cracks after they occur, has been rarely addressed. Considering that one of the primary objectives of self-healing concrete is to improve the ability of concrete to block water flowing through cracks [12,28], quantitative evaluation of rapid self-sealing of cracks by SAPs in terms of the change in the amount of water runoff through cracks over a relatively short period of time is considered extremely important. This is because rapid self-sealing of cracks by SAPs is expected to eventually improve the self-healing efficiency of cementitious materials in the long term.

Consequently, the purpose of this study is to quantitatively evaluate rapid self-sealing of cracks in cementitious materials containing SAPs. Mortar specimens were prepared with SAP dosage as a variable, and a through crack was induced in the specimens. A water flow test was performed, in which the change in the amount of water runoff through the crack over time was monitored as soon as the first ingress water was provided after crack occurrence so that the effect of swelling of SAPs on rapid self-sealing of cracks could be quantitatively evaluated. Furthermore, X-ray CT, cryofracture scanning electron microscopy (SEM), and optical microscopy were employed to visually capture the swelling behavior of SAPs before/after cracks occurred. In addition, the effect of SAP dosage on strength development in SAP-added materials was investigated.

2. Experiment

2.1. Materials

In this study, type-I ordinary Portland cement (OPC) (fineness: 3499 cm^2/g, density: 3.13 g/cm^3) and standard sand conforming to ISO 679 were used in the experiment. Table 1 shows the chemical composition of the OPC used in this study.

The swelling ratio of SAPs varies with factors such as the degree of cross-linking in SAPs, chemical structure of monomers constituting the SAP network, and properties of solutions, such as pH, ionic concentration, and temperature [29,30]. In addition, it is affected by the size of SAPs [30]. Spherical SAPs (polyacrylate-co-acrylamide, density: 1.38 g/cm^3) were used in the test. They were sieved using a wire-woven sieve conforming to ASTM E11. Fig. 1 shows the particle size distribution of SAPs in dry and fully swollen states in different solutions, measured using a particle analyzer (Model: Mastersizer 3000, Malvern Instruments Ltd.). The estimated number of SAP particles used in each measurement was approximately 17,000 and the SAPs in swollen state were measured at 10 min after SAPs were immersed in the solutions. The mean particle diameter of dry SAPs was approximately 252 μm, while SAPs swelled up to approximately 1700 μm in distilled water and 1010 μm in tap water. Considering a relative short immersion period of 10 min, it is expected that the swelling ratio of bulk-polymerized SAPs will be different from that of the SAP particles shown in Fig. 1, when measured using the particle analyzer used in the test [30].

Table 2 shows the mixture proportions of the mortar specimens used in the test. The water-to-cement and sand-to-cement ratios

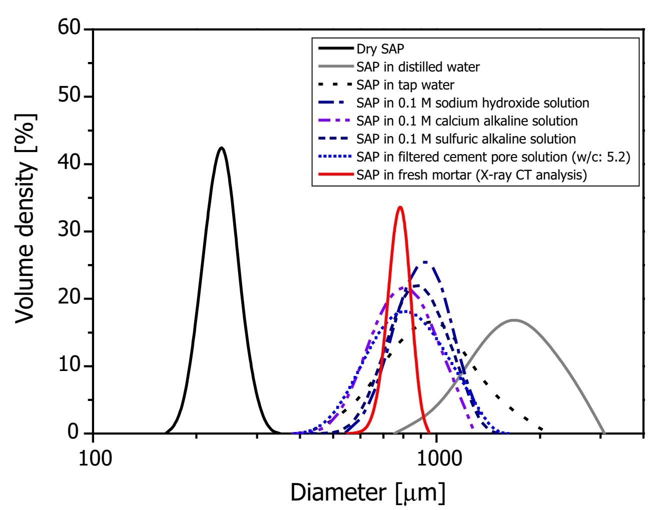

Fig. 1. Particle size distributions of SAPs used in the test.

Table 1
Chemical composition of OPC used in the experiment (%).

SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Na_2O	LOI*
20.5	4.97	3.02	61.8	2.71	2.35	0.72	0.33	2.36

* Loss of ignition.