



Influence of superabsorbent polymers on the surrounding cement paste



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ABSTRACT

In the last decade, the influence of superabsorbent polymers (SAP) on the performance of cementitious materials has been studied extensively. However, the formation mechanism and properties of the affected zone around SAP are still not well understood. Spherical SAP particles of a large size (1.5–2 mm) were used in this study to magnify the zone influenced by SAP. Hydration characteristics, capillary pore structure, microhardness and microstructure of the affected zone were investigated. The degree of hydration of the affected zone is higher than the unaffected zone by about 5%, an equivalent of an increase in w/c by 0.04. Capillary porosity is increased at early age, but reduced at later age, due to the desorption process of SAP. Results on the microhardness, the degree of hydration and the pore structure of the affected zone indicate a dense transition zone is produced in the presence of SAP.

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1. Introduction

In the past few decades, high-performance concrete (HPC) with a low w/c ratio ($w/c < 0.4$) has witnessed huge development thanks to the wide use of chemical admixtures and supplementary cementitious materials (SCM), such as the polycarboxylate superplasticizer and silica fume. Powers' model [1,2] suggests that cement could only hydrate completely with a w/c ratio above 0.42 [1]. The w/c ratio is generally above 0.38 for a normal concrete, and is between 0.20 and 0.38 for HPC [3]. Concretes with a low w/c ratio are very susceptible to cracking caused by autogenous shrinkage associated with the self-desiccation [4]. SAP are introduced into HPC as an internal curing agent, and the stored moisture in the SAP serves as water reservoirs which will be released during concrete hardening [5].

Benefits from the application of SAP in concrete generally include humidity regulation, shrinkage mitigation, cracking reduction, etc. According to a series of studies, performances of concrete are influenced considerably by SAP. For workability, it was believed that the plastic viscosity of concrete was increased by the presence of dry SAP particles, which would accordingly influence the rheological behavior of fresh concrete [6]. For shrinkage, it was suggested that SAP could reduce the plastic shrinkage [7], autogenous shrinkage [8] and drying shrinkage

[9]. For mechanical properties, SAP was often found to have a negative effect on the strength of concrete at early ages [10,11], whereas a higher strength at later ages was reported [12]. For durability, some studies showed that the introduction of SAP had almost no negative effect on the permeability [13,14], although a certain amount of pores were left in the hardened concrete by SAP. Furthermore, the freezing–thawing resistance of concrete was improved as a result of the increased void content [15]. However, to the knowledge of the authors, only a few studies [14,16–18] have been focused on the “affected zone” which is directly influenced by SAP in concrete. Characterization of the affected zone of SAP differs, depending on experimental techniques used [14]. Mechtcherine et al. [16] found that no differences were seen between the surrounding cement paste and the other matrix areas, by using backscattered electron (BSE) image. However, Lam [17] found a dark grayish rim which seemed like an interfacial transition zone (ITZ) around the SAP void, by using a BSE detector. Mönning [18] also detected an affected zone with a thickness of 60 μm around the SAP particle, by observing the altitude profile changes compared to the reference layer.

The release of entrained water from SAP would have a significant effect on the properties of cement paste, such as the hydration process, the pore structure evolution, etc. According to some recent studies, the primary hydration heat peak occurred earlier compared to the corresponding pastes with the same water amount but without SAP [19], the cumulative heat of hydration and the ultimate degree of hydration were increased because of the release of water retained in SAP [19–22]. After the water release, extra voids were created, and the total porosity of the whole cement paste was increased [16,21]. It was also

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reported that the water release continued the hydration of cement so that the porosity was decreased at late ages [23]. It should be noted that these existing studies were mostly focused on the whole cement paste with or without SAP, and were thus an overall representation. Investigations on the properties of the affected zone alone are still very few.

Reasons for the scarcity of research on the affected zone by SAP are mainly as follows: Firstly, the particle size of SAP usually used in concrete is a few hundred micrometers, thus the concomitant voids after water release of SAP are very small. This makes the potential ITZ around SAP very hard to distinguish. Secondly, some gel polymer SAP powders prepared after being dried by crushing produce irregular particles, thus producing irregular small voids in the concrete.

As a result, the present study used a large spherical SAP with a dry particle size of 1.5–2 mm which makes it more feasible to identify the affected zone [24]. Properties, such as hydration heat flow, degree of hydration, pore size distribution, microhardness and backscattered electron imaging of the affected zone around SAP and the unaffected zone were studied. It should be noted that the large size SAP which has been used is not practical for internal curing.

2. Materials and methods

2.1. Materials

Portland cement was used with a Blaine fineness of 360 m²/kg and the following chemical composition in wt.% (SiO₂: 21.70, Al₂O₃: 4.76, Fe₂O₃: 3.57, CaO: 63.01, MgO: 2.12, and SO₃: 1.98). Its physical and mechanical properties were shown in Table 1. A polycarboxylate-based high-range water-reducing admixture with 23.5% solid content was used only for the disc-shaped specimen preparation.

Polyacrylic acid-based SAP spheres were used with a dry particle size of 1.5–2 mm, and 4–5 mm after swelling. The maximum water absorptivity of spherical SAP in pure water was 115 g/g SAP (Fig. 1). Large spherical SAP particles used in this study have a lower water absorptivity than the SAP powders (100–200 μm in dry particle size) commonly utilized for internal curing (Fig. 1), but they have similar desorption behavior in the cement paste affected by osmotic pressure and humidity gradient [24]. As a result, SAP particles were pre-soaked before use. All the presoaked SAP spheres used in this study had almost the same particle size which meant the same amount of absorbed water.

2.2. Specimen preparation

To create the spherical pores in the cement paste formed by SAP, a hemispherical SAP particle was positioned at the bottom of a culture dish, then fresh cement paste was slowly poured in to form the disc-shaped specimen with a thickness of 15 mm and a diameter of 85 mm. The cement paste mixtures with w/c ratios of 0.24, 0.26, 0.28, 0.30, 0.32 and 0.34 (all the w/c mentioned in this paper means nominal w/c), are mixed in a standard 2.5 L mixer for 2 min with a revolution speed of 62 ± 5 r/min, and another 2 min with a revolution speed of 126 ± 10 r/min, according to the Chinese national standard GB/T 1346-2011. The spherical SAP was pre-soaked in solutions with given concentration which would be discussed later. When red dye was added into the solution, it could also be used to track the water diffusion trace of SAP. The disc-shaped specimen was then sealed with plastic films

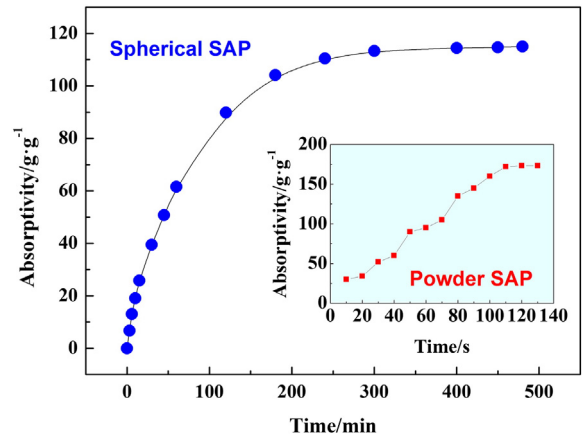


Fig. 1. Water absorption behavior of large SAP spheres and fine SAP powders in pure water.

to prevent moisture exchange, and cured at a temperature of 20 °C. The disc-shaped specimens were used to for the degree of hydration analysis, microhardness test, pore structure investigation and BSE imaging.

The disc-shaped specimen was shown in Fig. 2. The cement paste with SAP was divided into two zones: Section A and Section B. Section A means the affected zone close to SAP, and Section B indicates the unaffected zone which was not influenced by SAP. The following work mainly focused on the difference between the Section A and the Section B. The Section A and Section B could be distinguished by red dye, which was also described in [24].

2.3. Equilibrium concentration between SAP and cement paste

In this study, the spherical SAP particles were pre-soaked before mixing in the fresh cement paste. As we all know that the pre-absorptivity of SAP would affect the absorption/desorption behavior of SAP in cement paste dramatically. When the pre-absorptivity of SAP is lower than the equilibrium absorptivity (absorptivity of SAP in the pore solution), the SAP particles would absorb first and then release. When the pre-absorptivity is higher than equilibrium absorptivity, the SAP particles would always release. To simplify the original state of presoaked SAP spheres after mixing in the fresh cement paste, the SAP particles were presoaked with an equilibrium state so that the presoaked SAP would not absorb first or release too fast. In other words, the presoaked SAP would remain relatively steady until the concentration of pore solution goes up. Thus, the presoaked SAP spheres would only experience the desorption process, since this paper was focused on the effect of desorption on the zone around SAP.

The moisture exchange between SAP and solution is mainly controlled by osmotic pressure which is principally affected by the concentration of solution, as described by Eq. (1). In fact, the equilibrium state of presoaked SAP in the cement paste could be controlled by limiting the absorbing time if absorbed in distilled water, or using proper concentration if absorbed in solution, both to get the equilibrium absorptivity. In this study, sodium chloride solution with proper concentration was used to reach the equilibrium state in the cement paste. Solution with

Table 1
Physical and mechanical properties of cement.

Blaine specific surface (m ² /kg)	Surface density (g/cm ³)	Setting time (minute)		Compressive strength (MPa)	
		Initial set	Final set	3-day	28-day
360	3.01	130	210	35	62

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