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Permeability and pore structure of microcapsule-based self-healing cementitious composite

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HIGHLIGHTS

- Studied permeability of self-healing cementitious composites, including pore structure.
- Small amounts of microcapsules positively affect pore structure and impermeability.
- Self-healing efficiency increases with increasing microcapsule content and particle size.
- Series relationships were obtained to predict relationship of impermeability vs. pore structure parameters.

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ABSTRACT

The effects of adding organic microcapsules having various particle sizes and composition on the permeability, carbonation resistance, pore structure, and self-healing efficiency of cementitious composite material were studied using rapid chloride migration, water pressure penetration, carbonation, and mercury intrusion porosimetry tests. It was found that increasing microcapsule content to 3% improved pore structure and impermeability. However, with 6% microcapsule content, the material impermeability became even lower than that of a specimen without microcapsules. The variation of particle sizes had a slight negative impact on impermeability although it improved pore structure. Increasing microcapsule content and particle size enhanced impermeability, pore structure, and self-healing efficiency of specimens.

1. Introduction

Concrete is widely used in modern buildings owing to its excellent performance and relative low cost. However, most concrete inevitably cracks because of its weakness in tensile strength and under conditions of plastic shrinkage, thermal stresses, drying shrinkage, corrosion of reinforcement, weathering, settlement, loading during the plastic stage, or after hardening [1]. Unless these micro-cracks can be effectively repaired, these will continue to be detrimental to the performance and durability of concrete structures, which can lead to catastrophic failure [2]. In recent years, microcapsule-based self-healing cementitious composites, which autonomously detect and repair damage, partially or completely, have gradually aroused the interest of researchers [3,4].

The list of studies on self-healing behaviour in cementitious composite is summarized in Ref. [1], and a number of review papers [5–10] on the subject of self-healing materials have been published. There are many self-healing techniques including organic microcapsule-based self-healing, bacteria-based biological self-healing, superabsorbent polymers which can swell to block a crack from intruding fluids, and use of expansive agents and mineral admixtures, etc. [11–14]. Among them, organic microcapsule-based self-healing is regarded to be quite effective and exceptional in that it preserves the healing agent until damage triggers self-repair, which is indeed extraordinary from the perspective of durability and economy [15]. Self-healing ensues the moment a micro-crack is generated because the embedded microcapsules are broken along the crack, subsequently releasing the healing agents inside the microcapsules, which then mend the crack [16]. At present, although the microcapsule technology is not fully developed, its future widespread use, notably in concrete structures, is quite promising.

Recall that the permeability of concrete dramatically increases as soon as it cracks. The fissures, although ordinarily hairline, permit corrosive ions to invade and damage the concrete, which can eventually reduce its service life. In fact, many concrete structures inevitably degrade and deteriorate over time due to

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water permeation [17]. Permeability is also an important index to measure self-healing efficiency. Therefore, to obtain the best and most economical self-healing effect, and to minimize the harm of corrosive ion penetration, the study of the permeability of self-healing concrete is necessary.

There have been numerous studies on the permeability of concretes, mortars and pastes [18]. It was discovered that permeability changes with the change of pore structure and its development. Reference [19] indicated that three broad considerations influence the permeability of cementitious composites, namely: 1) factors that alter the initial pore structure, such as water-cement and mineral admixtures (e.g. silica fume, fly ash, and blast furnace slag) and additives (e.g., water reducing agents); 2) factors that affect the development of pore structure, such as curing, age, and binder activity; 3) surrounding, such as the driving force that causes water to flow.

Feldman elaborated on factors related to concrete permeability, presented a number of representative formulas for ionic permeability, and gave a broad description of the test method and development direction of pore structure [20]. A review of different pore structures and permeability test methods were presented, as well as their advantages and limitations in practical application [21]. Nevertheless, there remain very few studies on the combination of macroscopic properties and micro-properties of microcapsule-based self-healing cementitious composites, and the self-healing efficiency on the micro-structure level [10]. To this end, this study focuses on the permeability of self-healing concrete from the perspective of its relationship with pore structures.

In this study, microcapsules were synthesized by in situ polymerization and mixed into cement to produce microcapsule-based self-healing cementitious composites. The influence of average particle size and content of microcapsules on permeability and self-healing efficiency of self-healing cementitious materials were investigated by rapid chloride migration (RCM), water pressure penetration, and carbonation tests. Moreover, the effect of microcapsule particle size and content on a pore structure parameter and self-healing efficiency were investigated by mercury intrusion porosimetry (MIP) test. Based on experimental data, the relationships between pore structure, impermeability of cementitious composite, and healing rate were established.

2. Experimental program

2.1. Microcapsules

As mentioned earlier, the microcapsules were made by in situ polymerization. Epoxy resin was used as core enclosed in a urea-formaldehyde resin shell. The specific synthesis method can be found in [22].

The size of microcapsules can be adjusted by changing the stirring speed during the synthesis process, and in this research the stirring speeds were 200 rpm, 400 rpm, and 600 rpm, respectively. Regarding form, microcapsule shape was observed by scanning electron microscopy (SEM). It can be seen from Fig. 1(a) that the microcapsules are spherical and regularly shaped. The dimensions of microcapsules were also determined through SEM. Three hundred microcapsules were randomly obtained from samples that were prepared at different rotational speeds, and the average particle size d was calculated by formula (1):

$$d = \frac{1}{n}\sum_{i=1}^{n} d_i \tag{1}$$

where d_i is the size of the i-th microcapsule (μm); n is the number of microcapsules.9*//

The shell thickness of ruptured microcapsules was also measured by SEM, as shown in Fig. 1(b). The average particle size and shell thickness of the organic microcapsules prepared at different stirring speeds are summarised in Table 1, and particle size distribution is shown in Fig. 2.

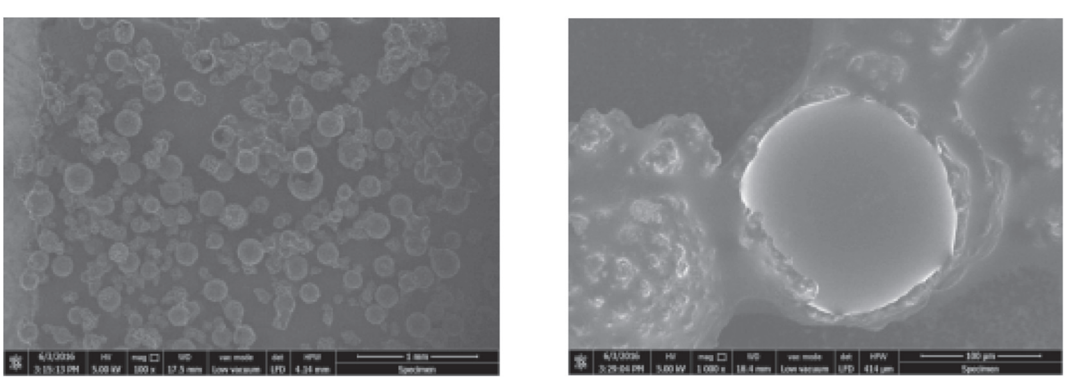
2.2. Preparation of specimens

Test specimens were made of the following materials: Portland cement PII42.5R type (Guangzhou Zhujiang Cement Ltd Co., ISO standards from Xiamen SO Ltd Co.); curing agent, MC120D (Guangzhou Chuanjing Electric Materials Ltd Co.) [23,24].

Three sizes of microcapsules (150.05 μm, 205.62 μm, 243.16 μm) and three percentages of microcapsule content (0%, 3%, and 6% of cement mass) were considered. Using these microcapsules, three types of specimens of different sizes were prepared for this investigation: cylindrical specimens of ϕ100 mm × 50 mm were used for RCM and MIP tests; conical frustum specimens were for water pressure penetration test, of which the upper surface diameter, the lower surface diameter, and height were 70 mm, 80 mm,

Table 1
Average particle size and shell thickness of microcapsules.

Stirring speed (r/min)	Average particle size (μm)	Shell thickness (μm)	Standard deviations of shell thickness
600	150.05	4.86	1.3
400	205.62	7.54	2.2
200	243.16	9.62	2.5



(a) Microcapsules (b) Shell thickness

Fig. 1. SEM images of microcapsules.