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Self-healing cementitious materials based on bacteria and nutrients immobilized respectively

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HIGHLIGHTS

- Bacteria and nutrients were immobilized respectively into ceramsite to improve the self-healing effectiveness of cementitious materials.
- The depth of cracks before and after restoration was investigated by staining the section surfaces of specimens.
- The flexural strength of specimens after repairing was studied to characterize the repairing effectiveness of cracks.
- The precipitation formed in cracks was analyzed by SEM/EDS and XRD.

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ABSTRACT

Small cracks in concrete may develop into large cracks to decrease service life of concrete structures. It is necessary to restrict the development of early age small cracks promptly. This study presents a bio-restoration method to improve the self-healing effectiveness of the cement-based materials cracks rapidly. Ceramsite carrier was used to immobilize bacteria, while substrate and nutrients mixed evenly were immobilized into other original carrier. The section surface of paste specimens before and after curing was investigated by staining. Water permeation coefficient and flexural strength test were applied to characterize the repairing effectiveness of specimens. Experimental results show that plenty of white precipitation generated on the section surface after curing 21 days and the apparent water permeation coefficient of specimens changed slightly after restoring 28 days. The area repair rate of section surface of the samples with bacteria and nutrients immobilized into ceramsite was run up to 87.5%. The flexural strength of specimens repaired could increase from 56% to 72% than other microbiological methods. SEM/EDS and XRD analysis results show that the precipitation formed in cracks is calcite.

1. Introduction

Concrete is one of the most widely construction materials in the world. However, with low tensile stress, it may cause minor cracks easily during the service life, which will degrade the performances of materials. And it will have a further impact on durability of concrete structures. Many methods such as repairing in time or self-healing for cracks could prolong the service life of them [1–5]. Since many researchers found that some bacteria could induce or improve the precipitation of calcium carbonate which called bio-mineralization, self-healing for cracks in concrete by precipitation of calcium carbonate has been developed for many years [6–9].

The effect on the deposition of calcium carbonate by a kind of anaerobic bacteria was first studied by Ramakrishnan et al. [9]. They found that carbonate produced with the decomposition of urea by microorganism. Calcium carbonate deposed by the Bacterial as nucleation site. This kind of mineralization induced by microorganism is widely used in the repair of cracks of cementitious materials [10,11]. Dick et al. [10] proposed that the cracks of limestone surface could be repaired completely by mineralization of urease. And the water absorption property of the limestone surface is greatly reduced after the restoration. Jonkers et al. [12] proposed that one type of aerobic bacteria could decompose specific substrate to $CaCO_3$. Then CO_2 is released in the process of reaction which can also trigger the precipitation of $CaCO_3$ in cracks. But the decomposed speed of substrate is slow and the quantity of CO_2 from substrate is limited. The research of Qian et al. [13] investigated the self-healing potential

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of early age cracks in cement-based materials incorporating the bacteria which could produce carbonic anhydrase. The results indicated that the bacteria showed excellent repairing ability to small cracks formed at early age of 7 days. However, calcium carbonate produced inside the cracks of cement-based materials is minimal because the calcium ion dissolve out from the materials slowly and the diffusion coefficient of carbon dioxide is small in water curing conditions [14]. Accordingly, calcium carbonate generated too insufficient to repair the cracks in depth. Besides, due to the harsh environment inside the concrete, microcapsules, ceramsite or porous expanded clay particle could been used to protect the bacteria [15–18]. Immobilization of bacteria into a protective carrier before adding them to the concrete is preferable [18]. Moreover, nutrients immobilized can be fully utilized.

This paper presents a bio-restoration method to improve the self-healing effectiveness of the cement-based materials cracks by immobilizing bacteria and nutrients respectively. We attempt to provide part of the carbon dioxide extra by yeast fermenting glucose. In our previous research, *Bacillus sphaericus* was found to be able to precipitate calcium carbonate rapidly. The equations is,

$$H_2O + CO_2 \rightarrow HCO_3^- + H^+$$

$$Ca^{2+} + OH^- + HCO_3^- \rightarrow CaCO_3 + H_2O$$

The new bacteria can promote the inter-conversion between CO_2 which from glucose and $CaCO_3$. That means CO_2 can be transferred to minerals precipitated in cracks when reacting with soluble Ca^{2+} rapidly. Finally, the self-healing effect of the cement-based materials cracks can be improved.

2. Materials and methods

2.1. Materials

Bacillus mucilaginous L 3 and *Brewers yeast* JCS 05 (China Center of Industrial Culture Collection, CICC) were used in this study. After incubated for 24 h in specific liquid medium, the fresh bacterial strain were harvested by high-speed (8000 r/min) centrifugation at 4 °C for 10 min. Then the bacterial strains were suspended in sterile deionized water. The final concentration of bacterial strain was about $10^8 \sim 10^9$ cells/mL.

The cement PII 42.5R was purchased from China United Cement Corporation (CUCC, Nanjing). Ceramsite carrier was used to carry the nutrients and bacteria. The component of the nutrient mainly includes sucrose, yeast extract and calcium nitrate. Glucose obtained from the Sinopharm Chemical Reagent Co., Ltd. was used for producing CO_2. The concentration of nutrient and glucose solution was 0.01 mol/L and 50 g/L respectively. Calcium nitrate obtained from Sinopharm Chemical Reagent Co., Ltd was analytically pure and diluted into 0.05 mol/L. Sudan Red III purchased from Sigma (St. Louis) was used to stain the section surface of the specimens before curing.

Table 1
Composition of the specimens in each series.

Group	Cement (g)	Water (g)	Ceramsite 1 (g)	Ceramsite 2 (g)	Ceramsite 3 (g)
C1	1200	504	146	0	0
C2	1200	504	0	146	0
C3	1200	504	0	0	146
C4	1200	504	0	73	73

Ceramsite 1 without any additions, ceramsite 2 with glucose only, ceramsite 3 with *Bacillus mucilaginous* and *Brewers Yeast*.

2.2. Preparation of the specimens

The ceramsite carrier was washed by water twice to clean dust and dried by drying oven. The bacteria solution contained *Bacillus mucilaginous* and *Brewers yeast* was mixed together. Then the ceramsite carrier with bacteria was put into the suction flask and filtrated for 1 h by vacuum pump. The nutrient, glucose solution was immobilized into the other ceramsite carrier by the same method.

Cement paste specimens were used for this study. All mixtures were designed with the water to cement ratio of 0.42. Four series of specimens were made and the composition of each series is shown in Table 1.

As shown in Table 1, C1 are the specimens with ceramsite 1 without any additions. C2 are the specimens with glucose only. C3 are the specimens with *Bacillus Mucilaginous* and *Brewers Yeast*. C4 are the specimens with nutrient and bacteria at the same time.

The ceramsite was mixed with cement and water to prepare prism specimens (40 mm × 40 mm × 160 mm) and cylinders specimens (φ, 110 mm; h, 45 mm). After casting, all molds were put in curing room with temperature of 20 °C and relative humidity of more than 90% for 24 h. The specimens were then de-molded and placed under the standard curing room with temperature of 20 ± 2 °C and relative humidity of 95% until the time of testing.

2.3. Cracks preparation and self-healing incubation

The pre-cracks of specimens from C1 to C4 were taken after curing 28 days. The cracks were formed on prism specimens by embedded method according to Luo et al. [19]. The prism specimens were wrapped with adhesive tape before bending test. The initial cracks were made by bending load which loaded to the prisms specimens until cracks penetrated the cross section. Then nails with different width were embedded into the initial cracks to achieve different width. The width of cracks was measured by Crack width measuring instrument (Beijing Koncrete Engineering Testing Technology Co., Ltd.) with accuracy degree of 0.01 mm. The treated cracked specimens were immerged in tap water with temperature of 30 ± 2 °C which was exposed to the atmosphere during the whole repair period.

2.4. Characterization methods of cracks self-healing

2.4.1. Water permeability test

The crack healing efficiency of specimens was evaluated by water permeation coefficient according to the method reported with modification [17]. The diagram of water permeation test is shown in Fig. 1. Firstly the cylinder specimen was casted into PVC mold so that the PVC mold could be connected to the PVC pipe. Before test, the joint was sealed tightly to avoid leakage. Then the water in PVC pipe was kept a fixed height to maintain a constant pressure on the surface of cylinder specimen. The volume of passed water could be measured easily during a period of time. Permeability coefficient of cylinder specimen before and after healing could be calculated according to Darcy's Law shown in Eq. (1). Where k is permeability coefficient, m/s; Q is amount of water flow, m^3/s; L is height of specimen, m; A is area of section, m^2; Δh is head difference, m.

$$k = Q \cdot L / A \cdot \Delta h \tag{1}$$

2.4.2. Flexural strength test

The flexural strength after repaired with different self-healing agent was tested according to the method reported by Li et al.