



# Self-healing of concrete cracks by use of bacteria-containing low alkali cementitious material

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## HIGHLIGHTS

- A new type of carrier based on the low alkali cementitious material was developed.
- The carrier was very effective in preserving the bacterial activity.
- Cracks up to 417  $\mu\text{m}$  was healed completely in 28 days by loading bacteria in the carrier.

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## ABSTRACT

Self-healing based on microbially-induced calcium carbonate precipitation has been proposed as a smart and environmentally friendly strategy for the repair of concrete cracks. It is advisable to incorporate bacteria-based healing agents in fresh state concrete during mixing. Although the selected bacteria are alkaliphilic spore-forming strains, they are still vulnerable to the harsh environment of concrete. In this paper, we developed a protective carrier for the bacteria by using calcium sulphoaluminate cement, which is a type of low alkali, fast hardening cementitious material. By regulating the composition of the carrier material and the content of healing agents, the compatibility of the carrier with both the healing agents and the concrete matrix was optimized. The carrier, which acted as a support for the bacteria, was effective in preserving the bacterial activity over a long period of time. After embedding this bacteria-based self-healing system in concrete, cracks up to 417  $\mu\text{m}$  with a crack closure near 100% was achieved in 28 days. Compared with plain mortar, the regain ratios of the compressive strength and water tightness increased 130% and 50%, respectively. The research suggests the potential application of this novel microbial self-healing system in extending the life span of concrete.

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## 1. Introduction

Since the crystal formation by bacteria as a general phenomenon was revealed almost half a century ago, it has been applied in a variety of fields such as geological engineering, oil industry, historic preservation, and civil engineering [1,2]. A series of applications involve plugging of rock system for oil recovery enhancement, consolidating and strengthening of sand columns, and protection of ornamental stones were achieved by bio-deposition or bio-cementation [3–10]. In most cases, calcinogenic bacteria were used that calcium carbonate ( $\text{CaCO}_3$ ) was produced during the process, which is known as microbially induced carbonate precipitation (MICP) [11–13]. Recently, this technique has been

explored for the crack remediation and durability improvement of concrete [14–18].

For concrete, a most used building material worldwide, cracking is almost unavoidable due to its inherent brittleness and the complex service environments. Although the presence of cracks may not impair the strength and integrity of the concrete structure immediately, it will reduce the durability of concrete because aggressive species like chlorides and sulfates can penetrate into the matrix through crack paths, particularly when a continuous network is formed by micro-cracks. Thus, timely repair work is needed. General repair methods often require periodical inspection and maintenance, which are quite laborious and expensive. Besides, repair agents are applied from outside that deep micro-cracks may not be possible to be healed due to the limit of penetration. In order to solve these problems, an alternative method based on self-healing process was proposed [19].

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As a matter of fact, concrete is capable of sealing micro-cracks in certain conditions, and it is known as “autogenous healing”. This phenomenon is primarily attributed to hydration of unhydrated cement particles or chemical precipitation of  $\text{CaCO}_3$ . Another category of self-healing is so-called “autonomous healing”. By simply adding specific healing agents in fresh state mixture, cracks will be healed from inside to outside in hardened concrete via the release of healing agents [20,21]. However, most studied healing agents are chemically based. Under such circumstances, self-healing of concrete by MICP was considered in the past decades. Compared with other types of healing agents, bacterially mediated  $\text{CaCO}_3$  is compatible with concrete and environmentally friendly. Up to now, three bacterial metabolic pathways associated with  $\text{CaCO}_3$  precipitation have been investigated for concrete crack self-healing. The first pathway is the enzymatic hydrolysis of urea, which is carried out by ureolytic bacteria [22–25]. The second one is the oxidation of organic carbon [26–30]. Another pathway is related to the denitrification process under anoxic conditions [31,32]. Among all the pathways of the precipitated bio- $\text{CaCO}_3$ , ureolysis-based type is easier to operate and control. It is also known for its high efficiency that the reaction rate of biogenic urea hydrolysis is approximately  $10^{14}$  times faster than the chemical rate [21].

Because of the alkalinity nature of concrete, alkaliphilic spore forming strains were used for self-healing. Bacterial spores, which are dormant state of cells, can withstand heat, dry, various chemicals, and have much longer shelf life. The prerequisite of self-healing is that spores together with relevant nutrients must be incorporated during casting. However, directly adding bacteria in matrix is inadvisable due to the harsh environment inside concrete. Jonkers et al. [27] reported that bacterial activity decreased significantly in the high pH (>12) environment and bacterial cells (1–3  $\mu\text{m}$ ) could be squeezed or crushed owing to the decrease of pore size (<0.5  $\mu\text{m}$ ) along with the hydration process of concrete. Therefore, encapsulation or immobilization of bacteria in a protective carrier is preferable. A variety of carriers have been studied previously. Jonkers et al. [33–35] proposed a self-healing system by loading bacterial spores and other bio-reagents in expanded clay particles. The maximum crack width that can be healed reached 0.46 mm after 100 days of incubation, compared with only 0.18 mm for the control. De Belie et al. [36–40] tested a series of carriers including hollow glass fibers, microcapsules, diatomaceous earth, silica gel, polyurethane, hydrogel, and granular activated carbon. Superiority of self-healing appears after several weeks incubation of directly immersion or wet-dry cycle curing. Compared with the reference specimens, the strength regain increased 60% while the water permeability decreased about 1–2 orders of magnitude. In our previous research, air voids were introduced by air entraining agents to accommodate bacteria [41]. The recovery of strength and modulus after bacterially-mediated healing was almost two times higher than that of control. Above all, a preferable protective carrier not only provide a shelter for bacteria, but has limited negative effects on bacterial activity and cement hydration.

In the present study, we tried to develop a bacterial healing system by encapsulating healing agents in calcium sulphoaluminate cement, a type of low alkali cementitious material. It is a fast

setting and hardening binder that often used for concrete repair. In this manner, the low alkali binder serves as a protective carrier which is compatible with concrete and not detrimental to the viability of bacteria. The aim of this study is to quantify the self-healing ability of concrete based on this system.

## 2. Materials and methods

### 2.1. Cultivation of bacteria

A ureolytic type bacterium, *Sporosarcina pasteurii* ATCC 11859, was purchased from the China General Microbiological Culture Collection Center (CGMCC). Bacterial strains were cultured in liquid medium according to the supplier's recommendation and supplemented with manganese to enhance spore formation. The medium contained 5 g peptone, 3 g beef extract, 20 g urea, and 0.01 g  $\text{MnSO}_4 \cdot \text{H}_2\text{O}$  per liter of distilled water. Urea was separately sterilized by filtration through a sterile 0.2  $\mu\text{m}$  filter and mixed with other ingredients which were autoclaved at 121 °C for 20 min. The pH was adjusted to 8 ~ 8.5 by using filter sterilized NaOH or HCl solution. Cultures were aerobically incubated at 20 °C on a water-bath shaker operated at 100 rpm. Growth was regularly checked quantitatively by an optical microscopy. The incubation was performed for over 14 days until more than 90% of cells were spores. The cells were harvested by centrifuging the culture (4000 rpm, 20 °C) for 10 min and resuspension in sterile saline solution (0.15 M NaCl), then subjected to a pasteurization process of 20 min in a 80 °C water bath in order to eliminate vegetative cells. The suspension of spores (about  $10^9$  spores/mL) was stored in a 4 °C fridge for future use.

### 2.2. Tests on encapsulation material

The calcium sulphoaluminate cement (42.5R, Qilin Co. Ltd, China) was used as the encapsulation material for bacterial spore loading owing to its nature of low alkalinity. Silica fume (Elkem Materials) was added in the amount of 0%, 20%, and 40% by mass of calcium sulphoaluminate cement to further reduce the alkalinity of the material. Table 1 shows the chemical composition of the cementitious materials. In order to evaluate the alkalinity of the encapsulation material, a binder solution was made by a large water-to-binder ratio (*w/b*) of 10 to ensure a thorough hydration. The calcium sulphoaluminate cement together with different amount of silica fume were mixed with water in a 100 mL falcon tube and put on a shaker at 100 rpm for 1 h. Then the mixture was filtered and the pH of the filtrate was measured by a pH probe (Mettler Toledo pH Meter Kit). For comparison, Ordinary Portland cement (P-O 42.5, Conch Co. Ltd, China) whose composition is shown in Table 1 was tested by the same procedure. Three replicates were tested in each series.

Nutrients include peptone and beef extract should also be loaded in the encapsulation material. The influence of the nutrients and bacterial spores on the hydration of calcium sulphoaluminate cement was investigated. At a certain spore addition (1 mL suspension of spores with concentration of  $10^9$  cells/mL), varied concentration of nutrients (0%, 1%, 1.5%, 2%, 3%, 4% in mass of calcium sulphoaluminate cement) were used. Table 2 shows the detailed

**Table 1**  
Chemical composition of the used cementitious materials.

Materials	CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	Fe <sub>2</sub> O <sub>3</sub>	SO <sub>3</sub>	MgO	K <sub>2</sub> O	TiO <sub>2</sub>	P <sub>2</sub> O <sub>5</sub>
Ordinary Portland cement	54.86	21.86	6.33	2.61	2.66	2.60	0.68	0.27	\
Calcium sulphoaluminate cement	43.00	8.28	33.36	1.95	7.90	1.69	0.23	1.35	0.14
Silica fume	0.29	93.80	0.26	\	0.45	0.56	0.54	\	\

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