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Healing cement mortar by immobilization of bacteria in biochar: An integrated approach of self-healing and carbon sequestration

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ABSTRACT

Self-healing of cracks in concrete by bacterial carbonate precipitation is an effective mechanism to ensure better serviceability of civil infrastructure. This study explores biochar, derived from wood waste, as carrier for carbonate precipitating bacteria spores in cement mortar to seal cracks, and recover strength and permeability of healed samples. Superabsorbent polymer (SAP) and polypropylene microfibers (PP) were added to ensure moisture availability to bacteria and control crack propagation during damage of mortar. Samples were damaged by pre-loading to different levels – 50% and 70% of peak strength at 14-day. Experimental results show that biochar immobilized spores combined with SAP and PP precipitate copious amount of calcium carbonate, which completely sealed cracks up to 700 μm. This mix also showed highest recovery of impermeability and strength under both levels of preloading. Improvement in strength by 38% and reduction in water penetration and absorption by 65% and 70% was observed by immobilization of spores in biochar, compared to directly added spores. From comparison between samples, it was found that inclusion of PP fiber contributed to recovery of strength and impermeability, while SAP ensured higher precipitation of bacterial induced carbonate precipitation. The study suggests that spores immobilized in biochar has potential to offer excellent self-healing in cement composites. Using biochar is also a carbon sequestration strategy because of high volume of stable carbon stored in biochar particles during pyrolysis. Therefore, the proposed material combination would offer carbon storage in buildings, while also promoting waste recycling.

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1. Introduction

It is well known that cementitous materials are susceptible to cracking due to various loading and environmental factors. Micro-cracks allow easy entry of foreign chemicals into the cementitous matrix, that leads to further damage and results in loss of stiffness and strength of the structure [1,2]. Therefore, structures made with cement mortar or concrete need repair and maintenance operations, that are often cost intensive and add to environmental pollution related to construction industry. For example, according to an estimate crack repair and maintenance incur cost of $147 per m^3 of concrete, despite the production cost per m^3 of concrete ranges between USD 65 to USD 80 [3]. In general, manual repair operations that often rely on spraying or injecting of chemical sealants suffer from several limitations that include material incompatibility, low thermal resistivity, expansion and delamination besides raising concerns of human toxicity during their service life and disposal [4,5]. Therefore, over the past decade, bio-based self-healing has received considerable attention as a more sustainable and eco-friendly mode of crack repair in cementitous materials. While manual repair of cracks is difficult in cases of limited accessibility to the cracked zone, bio-based self-healing techniques can seal cracks and voids from interior without manual intervention.

Self-healing by certain bacteria species, primarily of genre Bacillus involve sealing of cracks by precipitation of calcium carbonate by direct precipitation or ureolytic decomposition of calcium containing compounds including calcium nitrate and calcium lactate [6–9]. Although genre Bacillus is alkaliphilic and their spores can lie dormant in alkaline environment without food for many years, the metabolism of the bacteria cells are affected by high pH ($pH > 11$) and dry environment [8]. For example, Jonkers et al. [6] noted dramatic reduction in viable spore after 22 and 42 day in

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concrete when spores of Bacillus cohnii was directly added to concrete mix. However, when spores were immobilized in lightweight aggregate, the viability spores were unaffected up to 6 month age of concrete [10]. It indicates that immobilization of bacteria spores is essential for their desired activity in dry and alkaline environment of cement composite for longer period of time.

In the past, several carrier material for bacteria spores have been investigated, including lightweight clay aggregate (LWA) [10], diatomaceous earth (DE) [8], polyurethane and silica gel [11], hydrogel [9], graphene nano-platelet [7], zeolite [12] and polymer microcapsules [13]. However, the technique of micro-encapsulation, including poly condensation technique is novel, yet complex and expensive to be rolled out on a wide scale for industrial application. It means that there is a need for a better immobilization technique of bacteria spores, that would not significantly affect mechanical properties of self-healing cementitous composites, while offering improved self-healing compared to control.

Use of porous LWA provided protection to bacteria and healing of cracks with width up to 0.46 mm was observed besides high recovery of water permeability. However, the major limitation of using LWA to partially replace hard granite aggregates is the significant reduction in strength (up to 50% at 28 day) [10], which may not satisfy the structural requirement. DE was explored as an immobilization means for bacteria spores in mortar. However, most of the pores of DE is in the range of 0.1–0.5 μm, while the spore size of bacteria ranges from 1 to 2 μm [8]. It means that the spores were absorbed onto the surface of DE and higher quantity was needed to increase survivability of spores. The limitation of mixing high amount of DE in cementitous mix is the reduced workability because DE tends to absorb high amount of mixing water that affect flowability of the fresh cement paste.

Khaliq et al. [7] reported that graphene nano-platelet (GNP) and porous LWA can offer protection to bacteria spores (B. Subtilis) and distribute them uniformly for self-healing of concrete cracks. Bacteria immobilized in GNP showed high self-healing when samples were pre-cracked at early stages (3 day and 7 day), while bacteria soaked in porous LWA was more effective for sealing cracks, that are introduced at late age of concrete. Porous LWA is more efficient than GNP in holding part of mixing water during concrete preparation, that is later provided for bacterial action.

Bhaskar et al. [12] demonstrated that zeolite can act as immobilization medium for bacteria spores (Sposarcina ureae and Sporosarcina pasteurii) to generate self-healing effect in normal and PVA fiber-reinforced mortar. Sorptivity after healing was reduced by 65% and 50% after healing in normal and fiber reinforced specimens with zeolite immobilized bacteria. Higher reduction in sorptivity in PVA fiber reinforced mortar was attributed to crack bridging effect of fiber that restricted the crack width, thereby achieving more complete sealing.

Similar findings were reported by Luo et al. [14] - repair rate of crack reduced drastically with increase in crack width. About 85% sealing was observed when crack width was below 300μ. Moisture is essential for respiration of bacteria and metabolic conversion of substrate to calcium carbonate. Luo et al. [14] reported that highest sealing was observed in case of damaged specimen subject to water curing and wet-dry cycle, which is also supported by Wang et al. [13]. It is clearly understood from the existing studies that higher healing can be obtained if crack width is limited and there is supply of moisture for bacterial action.

Therefore, this study proposes the use of biochar, produced from organic wastes (wood in this case) as a novel carrier material for bacteria spores in cement mortar. Some of the most favorable properties of biochar that make it appropriate as a construction material are its low thermal conductivity, high chemical stability, and low flammability [15]. Depending on pyrolysis condition, biochar contain pores with size ranging from 2 nm to 20 μ that can also house the bacteria spores within the cement mortar and help in uniform distribution within the hardened mortar. The concept is similar to use of biochar as soil enhancer and a material to improve plant growth by providing a shelter for rhizobium bacteria and other soil bacteria species [16,17].

Moreover, biochar has received attention as a potential material candidate to address climate change because it sequesters high volume of stable carbon in its structure, which is inherited from the feedstock itself. Biochar has the potential of reducing net greenhouse gas (GHG) emissions by about 870 kg CO_2 equivalent (CO_2-e) per tonne dry feedstock, of which 62–66% are realized from carbon capture and storage by the biomass feedstock of the biochar [18]. With sprawling urbanization in developing countries around the world, use of biochar as an additive and carrier for spores in self-healing infrastructure would provide an opportunity for high volume carbon sequestration that would turn cities into carbon sinks.

In addition to biochar, combination of superabsorbent polymer and polypropylene micro-fiber is used. It involves two mechanisms. The first mechanism is to achieve controlled cracking and increase fracture toughness through bridging action during micro and macro-cracking of the matrix [19]. Results reported by Bhaskar et al. [12] shows that use of polymer (PVA) fiber offered excellent crack bridging and increased self-healing efficiency. The second mechanism is to ensure supply of moisture to the cracked site over long term for bacterial metabolism. Superabsorbent polymers have the ability to absorb high volume of moisture from the surrounding and retain within its structure without dissolving [20]. The retained moisture by SAP is then desorbed under humidity gradient to provide for bacterial action. Moreover, studies show that SAP itself can heal cracks by improved autogenous healing or by swelling that blocks the crack path [19,21]. The aim of the current research is to investigate sealing of cracks and recovery of strength and permeability properties by combination of biochar (carrier for spores), SAP and PP microfiber.

2. Materials and methods

2.1. Micro-organism

Bacillus Sphaericus was chosen for this study because of its ability to form spores and survive in alkaline environment [9,11,13]. The medium used to grow the bacteria cells comprised of urea and yeast extract at 20 g/l of the solution. The size of the spores ranged from 2 μm to 4 μm.

Concentration of bacteria cells was determined on the basis of optical density by spectrophotometry [7]. The medium used to grow bacteria solution was selected as reference, which was subjected to spectrophotometer with a selected wavelength of 600 nm. Subsequently, 1 ml of medium solution containing bacteria spores was also subjected to spectrophotometer at same wavelength. The concentration of spore was determined using the method applied by Ramachandran et al. [22]. The concentration of cells in the final solution was found to be 10^{10} cells/ml.

The bacteria spore solution was immobilized by mixing with dry biochar powder in falcon tubes (15 g biochar + 30 ml spore solution). The falcon tubes were put in a shaker for 1 h at 100 rpm for sufficient soaking of spores into the biochar pores.

2.2. Biochar

The biochar used in this study was prepared by pyrolysis of mixed wood saw dust (soft wood) at 500 °C, and 10°C/min (slow pyrolysis). Saw dust is chosen as the feedstock because it is

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