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Review

Autonomous healing in concrete by bio-based healing agents – A review

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HIGHLIGHTS

- Effectiveness of self-healing by bacteria in concrete is discussed.
- Encapsulation techniques and materials for bio-based healing action is presented.
- Six robustness factors for effective healing by bacteria in concrete are highlighted and discussed.
- Assessment methods for bio-based self-healing are discussed and compared.

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ABSTRACT

Crack formation in concrete structures is inevitable due to deterioration throughout its service life due to various load and non-load factors. Therefore, repair and maintenance operations are needed to prevent cracks from propagating and decrease the structures' service life. However, accessibility to cracked zone may be difficult; besides such operations require capital and labor and contribute to pollution due to anthropogenic activities and usage of more repair materials. Self-healing may be a possible solution to reduce manual intervention. Autonomous crack sealing by bacteria induced carbonate precipitation is an environmental friendly mechanism which is studied intensively by many researchers worldwide. This review focuses on evaluation of crack healing by bacteria when it is added directly to the concrete or added after encapsulating it into a protective shell. Four key aspects that determine effectiveness of bacterial self-healing have been highlighted and discussed; they are capsule material and encapsulation of bio-agents, survival of capsules during concrete mixing, effect of addition of bio-agents or capsules on concrete properties, and sealing ability and recovery of mechanical and durability properties. Finally, research gaps and scope of future research work are identified and discussed.

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1. Introduction

Cracking is inevitable in concrete due to its relatively lower tensile strength and action of different load and non-load factors. Sources of cracking may be varied including plastic shrinkage, drying shrinkage, thermal stresses, external loading and rebar corrosion or coupled effect of multiple factors. For example, micro-cracks may form due to shrinkage but may propagate at a lower stress level when external loading is applied causing network of cracks. Such network of cracks gives easy access to moisture and chemicals to seep into the structure or degrade concrete chemically. Such problems are worse in the tropics due to higher moisture content in the air and high precipitation. Cracks can be manually repaired but there are several problems associated with manual maintenance and repair operations such as impact on environment, accessibility and cost. Different chemical and cement based repair materials are used presently. Cement production is associated with about 7% of global anthropogenic CO₂ emission [1] while chemical healing agents present threats including material incompatibility, health and environmental hazards [2]. Micro-cracks may originate in concrete just after construction or at advanced stage which is often unnoticed until it leads to major durability or structural issues. It is also a financial burden to carry out routine repair operations on facilities. Therefore, there is a need to find a sustainable way of healing cracks which involve less cost and eliminate the need of manual intervention.

Self-healing is an emerging concept of delivering high quality materials combined with the capability to heal damages and it has received much attention in past decade for application in building structures. Therefore, an effective self-healing mechanism may be able to reduce repair and maintenance works substantially and concomitant environmental and economic impacts. Recently, a sustainable mechanism of self-healing using microbial induced precipitation of calcium carbonate has been intensively studied to seal and heal cracks. Self-healing by microbes involve precipitation of calcium carbonate in cracks by direct action of bacteria species including *Bacillus Subtilis* on calcium compound such as calcium lactate [3] or by decomposition of urea by ureolytic bacteria such as *Bacillus Sphaericus* [4,5]. Calcium carbonate precipitation by microbes is compatible with concrete and the process of formation is environmentally friendly [6]. *Bacillus Sphaericus* is known to be harmless to human [7]. Additionally, during the process oxygen is consumed and therefore it also reduces the chance of reinforcement corrosion. The genre *Bacillus* has qualities including tolerance to high alkaline environment and moisture and capacity to form spores which make it suitable to use as self-healing agent in concrete. Therefore, *Bacillus* has been most commonly used in research studies as the bio-agent for calcite precipitation [3–5,8–10].

The review aims at evaluation of self-healing in concrete by biological action when the bacteria are directly added to concrete or added after encapsulation in mineral or chemical compounds. The discussion is conducted in the light of several key criteria including capsule material and encapsulation of bio-agents, survival of capsules during concrete mixing, viability of bacteria, effect of addition of bio-agents or capsules on concrete properties, and sealing ability and recovery of mechanical and durability properties.

2. Bio-based self-healing-mechanisms and approaches

Mentioned by Hammes and Verstraete [11], precipitation of calcium carbonate in natural environment is ideally influenced by concentration of calcium ions, pH of the solution, concentration of dissolved inorganic carbon and availability of nucleation sites. While the first three conditions relate to concrete matrix, the fourth one is provided by the bacterial cell itself. Bacterial precipitation may be achieved through different pathways like conversion of calcium compound such as Ca-lactate or hydrolysis of urea by (ureolytic) bacterial metabolism. In the first mechanism, crack openings let oxygen penetrate inside concrete and bacteria along cracked surfaces convert calcium lactate into calcium carbonate and carbon dioxide. If there are portlandite particles in the vicinity reaction with produced carbon dioxide would yield more calcium carbonate which may as well be used for healing. Therefore, it may be understood that this mechanism would be more efficient in case of fresh concrete when there are still unhydrated calcium hydroxide particles. The second pathway is precipitation of calcium carbonate through hydrolysis of urea into ammonium and carbonate. Bacteria *Bacillus Sphaericus* produce enzyme urease which act as a catalyst in the process. Negatively charged bacterial cell draws calcium ion from a calcium source such as calcium nitrate to react with the produced carbonate to precipitate calcium carbonate.

Therefore, in a nutshell, self-healing by bacteria may be accomplished by any of these mechanisms but the efficiency of healing in concrete would depend on a number of other factors including availability of moisture, crack area or width to be healed, age of concrete and survival of bacteria in long term.

2.1. Healing by direct addition of bio-agents to concrete

Jonkers and Schalngen [12] tested the feasibility of application of bacteria spores for development of self-healing concrete. Three species of bacteria mixed in cement stone were tested – *Bacillus cohnii*, *Bacillus halodurans* and *Bacillus pseudofirmus*. Cement stone chips were cured in yeast extract and peptone based medium and then tested for compressive and tensile strength. There was no significant difference between bacteria containing samples and control samples. Scanning electron microscope (SEM) images showed precipitation of calcium carbonate crystals after 12 days of incubation. However, in this preliminary research the organic carbon sources or food for bacteria were externally supplied and germination was observed once samples were cured in the medium. Possible effect of addition of yeast extract and peptone along with mineral precursor compound was carried out in a later study by Jonkers et al. [8]. For self-healing action to be accomplished, the primary challenge is the survival of bacteria in highly alkaline environment of concrete. Jonkers et al. [8] investigated the potential of carbonate precipitation by species of bacteria which can remain viable while being added directly to concrete. Two species of spore forming alkaliphilic bacteria - *Bacillus pseudofirmus* and *Bacillus cohnii* were tested. Calcium acetate and calcium lactate were tested as mineral precursor compound. It was observed that only calcium lactate did not affect strength and resulted in slight increase of strength. 20–80 μm sized calcium carbonate crystals were observed on specimens but the precipitate was observed only

in young samples (7 day) and was absent in 28 day samples. This could be due to reduction in viable spores of bacteria due to alkaline environment of concrete and reduced pore size which is also supported by Luo et al. [13]. However, it is claimed that this mechanism is better than urease based approach because it does not produce excess ammonia which may cause reinforcement corrosion or form nitric acid by further metabolism which may degrade concrete matrix.

Luo et al. [13] studied the effect of crack width, curing condition and age of cracking when alkali resistant spore forming bacteria were added directly to concrete along with substrate. Crack widths of 0.1–0.5 mm were introduced and three different post-cracking incubation conditions including wet curing, water curing (immersion in water) and wet-dry cycles (all at 25 °C) were tried. Healing was measured by area repair rate at different cracking ages of 7, 14, 28, 60 and 90 days. It was observed that under immersion in water condition crack width up to 0.3 mm were completely healed and healing ratio of cracks between 0.1 and 0.3 mm were about 85%. In terms of curing conditions, water curing and wet-dry cycles showed best healing performance although the repair rate was slower for wet-dry cycle condition. Early age cracks were repaired efficiently while cracks at later age were not as efficiently healed. This is attributable to low survival rate of bacteria in absence of any protective shell and drop in concrete porosity resulting in shorter transport distance of the mineral [13].

For efficient repair, it is important to study the properties of mineral precipitate as well as its bonding with the original matrix. Xu and Yao [14] studied mechanical performance of self-healing concrete incorporating non-ureolytic bacteria as part of mixing water and employed a nano-indentation technique to evaluate nanomechanical properties of the precipitated calcium carbonate and its bonding with parent concrete. The approach was also innovative because it incorporated basalt fibers, silica fume and air entraining agents to maintain integrity of samples while introducing cracks, reduce alkalinity of cement paste and introduce voids for survival of bacteria respectively. It must be noted that although introduction of air voids can be acceptable in temperate countries to improve resistance of concrete against freezing and thawing, it may be difficult to accept in the tropics because it may affect mechanical and durability properties. However, some improvement in fire resistance of concrete may be realized because voids allow for the release of vapour pressure within the concrete during fire. Calcium lactate and calcium glutamate were tried as mineral precursor compound. It was observed that conversion of calcium ion to carbonate was higher for calcium glutamate and the crystal formed had granular shape unlike those formed from lactate. However, the bonding of carbonates formed from glutamate and lactate were not significantly different although transition zone in case of carbonate from glutamate was thicker and denser than that from lactate. Better healing in terms of recovery of flexural strength was obtained in samples containing calcium glutamate. Crack widths varied from 0.1 to 0.4 mm and were partially sealed by mineral precipitates which are formed by bacterial metabolism. Specimens with bacterial spores displayed superior healing to control samples but its effectiveness was less than that of external healing. The main reason for such observation was limited supply of nutrients. Although voids did accommodate bacteria, their action was limited due to lack of nutrients.

Although appreciable self-healing efficiency has been demonstrated by axenic spores of bacteria, the main hindrance to the application lies in the cost of production of such cultures. Silva et al. [15] made an effort to produce a cheaper form of bio-agent which is in fact a microbial community called CERUP (Cyclic EnRiched Ureolytic Powder). The microbial source was a substream of a vegetable plant and CERUP was prepared under non-sterile conditions which resulted in cheaper production cost. Self-

healing effectiveness was measured by addition of both autoclaved and non-autoclaved CERUP over 4 weeks in terms of crack closure. Non-autoclaved CERUP demonstrated crack healing up to 0.45 mm width while for autoclaved CERUP, the maximum crack width healed was little lower at 0.37 mm. Although the production cost of such non-sterile healing agent is substantially lower, more tests like recovery of strength and durability need to be conducted to present a better idea of true self-healing.

2.2. Healing by addition of encapsulated bio-agents

Efficient self-healing would be realized when the sealing is effective over long term and can be performed throughout the lifetime of a structure. Therefore, survivability of bacteria is very important. However, when bio-agents are directly added to concrete there may be several barriers to survivability of bacteria. Jonkers [3] noted that even though bacteria spores were added lifetime of the unprotected spores was limited to only two months and therefore effective self-healing was observed only in young samples. There may be several reasons for this including alkalinity of cement matrix, mixing of concrete and hydration of cement. Activity may be substantially decreased when spores are exposed to high alkaline environment over long time. Moreover, during mixing some spores may be damaged due to mixing force or impact of aggregates. Hydration of cement reduces porosity and pore size of matrix over time to as much as 0.5 μm while bacteria cells are typically bigger than such size [4]. Therefore, with shrinkage of pores germination of cells may be drastically reduced or stopped at late stage of a concrete structure. One way to overcome this limitation is to encapsulate the bacteria to protect them without affecting any of concrete properties and carbonate precipitation by bacteria. Different encapsulation techniques to protect bacteria have been used in literature including expanded clay aggregate [3,16], diatomaceous earth [9], silica gel and polyurethane (PU) in glass tubes [5], melamine based microcapsules [4] and hydrogel [10,17].

2.2.1. Encapsulation in polymeric microcapsules

Wang et al. [4] used melamine based microcapsules to encapsulate spores of *Bacillus Sphaericus*. Calcium nitrate was used as the mineral precursor and added to the concrete during mixing along with other nutrients such as yeast extract and urea. The healing by bacteria was measured in terms of ratio between healed crack area and initial cracked area. Concrete with encapsulated spores showed healing ratio between 48% and 80% whereas for specimens without spores the healing ratio was limited to only 50%. Highest reduction in crack area was observed when specimens were subjected to wet and dry cycle with water as medium. The wet cycle was considerable longer, about 16 h which may be difficult to attain under normal conditions without manual intervention. Maximum crack width healed was about 970 μm . Capsule dosage of 3% performed best in terms of recovery of water permeability and crack sealing although less variation was observed in case of 5% dosage. However, it may be said that 3% dosage was optimal because reduction in concrete strength was higher in case of 5% compared to 3%.

2.2.2. Encapsulation in special cement additive

Use of hydrogel for encapsulation of bacteria is an innovative effort by Wang et al. [10] because this technique provides a source of internal moisture for bacterial activity and growth with least manual intervention. It also acts as protective shell for bacteria. Significantly higher healing, about 40%–90% was observed when bacteria were encapsulated together with bio-reagents in hydrogel. A maximum decrease in water permeability of about 68% was observed. Efficient healing by use of hydrogel encapsulated

bacteria is possible due to water uptake and retention capacity of hydrogels. Wang et al. [10] noted that pure hydrogel used could retain 70% and 30% of water absorbed after 12 h and 24 h respectively when exposed to air with 60% RH and 20 °C. It means that for tropical places which are characterized by high humidity and precipitation, hydrogels may be a good choice for absorbing and retaining water to support bacterial action. However, it also depends on type of hydrogel – ionic or non-ionic [10]. Ionic hydrogels are pH responsive and therefore their uptake capacity may be affected by chemicals present in air. Non-ionic may be better for such application since ions present in moisture does not affect its water uptake and retention capacity. In Wang et al. [4] where melamine based microcapsules were used, best healing performance was observed when wet and dry cycles (wet period of 16 h and dry period of 8 h) were used as curing condition. Using hydrogel, contact time with water may be reduced drastically. Additionally, there is little need of providing water manually as hydrogels are capable of absorbing moisture from air which opens up better prospects to use such concrete for actual construction.

In a recent study, Wang et al. [17] tested the feasibility of using modified sodium alginate based hydrogel as bacterial spore carrier. Bacteria spores were found to be viable after encapsulation in the hydrogel measured by oxygen consumption at damaged site in the concrete specimens. Some leakage of spores from the modified hydrogel was recorded during mixing of concrete although 99% of the spores remained intact in encapsulated form. Addition of 0.5% and 1% by mass of hydrogel did not significantly affect workability of concrete; however tensile strength and compressive strength deteriorated by about 23.40% and 30% respectively with addition of 1% of the hydrogel. Such adverse effect may be attributed to formation of macro voids due to addition of hydrogel. SAP has two opposite effects on concrete; while it supplies moisture for internal curing and help in strength development, it also generated voids in concrete and reduces strength. Dominance of these two effects depends on water-cement ratio (w/c), dosage of SAP and age of concrete [18]. Applying gel-space ratio concept, at w/c greater than 0.45, SAP addition has very less impact on strength development and may reduce strength. Wang et al. [17] used relatively high w/c (0.50) which may also be one of the reasons for reduction in strength in SAP added samples.

2.2.3. Encapsulation in lightweight concrete aggregate

In earlier research, Jonkers [3] used porous expanded clay aggregate to immobilize bacteria spores and calcium lactate which is the precursor compound for mineral precipitate. Clay aggregates are lightweight and soft and when they rupture, calcium carbonate is precipitated by bacterial action in presence of air. Samples tested after two weeks of immersion in tap water, showed maximum healed crack width of about 0.46 mm. After 6 months, no loss in viability of bacteria was reported so far. Clay aggregates have been used widely to develop lightweight concrete and they also act as internal source of moisture that is necessary to support bacterial precipitation action. However, a number of parameters such as amount of water in aggregate, expanded clay aggregate spacing and pore structure influence its efficiency [19]. The main limitation of using clay aggregates to replace normal granite aggregates is the consequent strength reduction. In normal strength concrete, aggregates form the bulk of concrete and aggregate toughness determines compressive strength of concrete. Tough aggregates in normal concrete resist cracks which eventually pass through the matrix which is relatively weaker than the aggregate. However, when clay aggregates are used, the cracks would most likely rupture the aggregates because aggregate toughness may be lesser than that of matrix. It means that soft aggregates introduce a weak plane and draw the cracks towards them. Due to introduction of lightweight clay aggregate Jonkers [3] observed as much as 50%

strength reduction at 28, which may not be acceptable for structural applications.

2.2.4. Encapsulation in special mineral compounds

Diatomaceous earth (DE) is a type of mineral compound rich in silica and formed from shell of microorganisms called diatoms. It has highly porous structure and therefore offers potential for immobilization. Wang et al. [9] used DE to immobilize *Bacillus Sphaericus*. Upon cracking once the bacteria are activated by air or water it would hydrolyze urea and precipitate calcium carbonate from the calcium nitrate that is used as mineral precursor. Crack width healed also depended on immersion medium (water or nutrient medium); however cracks with width between 0.15 and 0.17 mm were found to be almost completely healed. Higher amount of DE means better protection because bacterial cells are primarily absorbed in some of the hollow pores. However, DE has high fineness with many nano-scale pores. This leads to the drying up of mortar when high concentration of DE is used which absorbs moisture in the cement matrix.

Bacteria species and encapsulation material that have been used in bio-based self-healing application have been summarized in Table 1 along with the key findings from these studies.

3. Evaluation of bio-based self-healing system

In general, ensuring efficient self-healing action would require optimization and evaluation of different aspects like effect of bio-agents, capsule and overall system design [21]. Key aspects covered in this review are

- Capsule material and encapsulation of bio-agents.
- Survival of capsules during concrete mixing.
- Effect of addition of bio-agents and capsules on concrete properties.
- Sealing ability and recovery of mechanical and durability properties.

3.1. Capsule material and encapsulation of bio-agents

As mentioned in the literature wide range of materials may be used for encapsulation such as polymers, glass, diatomaceous earth, and clay aggregates. Glass is commonly used as the material for cylindrical/ tubular capsules: Wang et al. [5] to encapsulate bio-agents immobilized in polyurethane and silica gel. Glass capsules are available commercially and may be made easily to the desired length and diameter as required. It is comparatively easy to inject or pump in bio-agents in glass capsules. Also, clay aggregates used by Jonkers [3] are available commercially but the encapsulation or loading of bio-agents have to be done separately.

Clay aggregates were used by Jonkers [3] and Wiktor and Jonkers [16] to embed a two component healing agent consisting of calcium lactate and spores of *Bacillus*. Clay aggregates were impregnated in steps - twice with calcium lactate and yeast extract and a final step with *Bacillus* spore suspension. The impregnation steps were performed under vacuum condition and after each step the clay particles were dried at 37 °C in oven for 5 days. Wang et al. [10] introduced a novel technique of encapsulation by using hydrogel to encapsulate spores and nutrients. The hydrogel was first prepared in the form of sheets by mixing a polymer solution based on co-polymer of ethylene oxide and propylene oxide with an initiator followed by exposure to UV radiation. Sheets were subjected to freeze grinding to obtain fine powders in the size range of 500 µm. For encapsulation, spores were added to the polymer solution and the same steps as that of the manufacture of hydrogel were followed. Likewise three types of encapsulated materials

Table 1
Review of bacterial species, capsule material used and their functions in self-healing.

Species of bacteria used	Encapsulated (Capsule material)	Directly added	Mechanism	Major findings	Reference
Spore forming bacteria (species not mentioned)		X	Not mentioned in the study	a) High early healing was observed by water curing b) Higher the cracking age, lower is the extent of healing	[13]
Bacillus		X	Decomposition of calcium source to precipitate carbonate	a) Calcium source affects healing ratio- calcium glutamate performs better than lactate b) Bacteria remained viable for 4 months	[14]
Bacillus cohnii	X (Clay aggregates)		Metabolic conversion of calcium lactate	a) Crack width of 0.15 mm with length 8 cm completely sealed b) No loss of viability up to 6 months	[3]
Bacillus Sphaericus	X (immobilized in PU and silica gel inside glass)		Ureolytic decomposition of calcium nitrate	a) PU immobilized bacteria specimens showed lowest permeability b) Higher bacteria activity in silica sol c) Higher strength recovery in case of PU immobilization	[5]
Bacillus Sphaericus	X (Diatomaceous earth)		Ureolytic decomposition of calcium nitrate	a) Highest reduction of water absorption was observed in bacteria containing specimen b) Dosage of DE must be carefully adjusted because it causes loss in concrete workability	[9]
Bacillus Sphaericus	X (Melamine based capsules)		Ureolytic decomposition of calcium nitrate	a) Crack healing ratio of 48% to 80%; highest crack width healed is 970 μ m b) Permeability recorded for bacteria specimen is about 10 times compared to control c) highest reduction in crack area in case of wet-dry cycle	[4]
Bacillus Sphaericus	✓ (hydrogel) – one component (only bacteria) and two component (bacteria and nutrient) system		Ureolytic decomposition of calcium nitrate	a) Maximum crack sealing of 500 μ m under wet-dry cycles b) Permeability decrease of 68% for specimens containing hydrogel encapsulating both bacteria and nutrients together	[10]
Bacillus Sphaericus	✓ (Sodium alginate based hydrogel)		Ureolytic decomposition of calcium nitrate	Bacterial activity was observed only for encapsulated samples at crack face measured by oxygen consumption	[17]
Bacillus Subtilis	✓ (Lightweight aggregates and graphite nano-platelets)		Decomposition of calcium lactate	a) Bacteria can be distributed uniformly in concrete when immobilized in graphite nano-platelets (GNP) due to fine particle size and uniform dispersion of GNP b) Bacteria immobilized in GNP showed high self-healing when samples were pre-cracked at early stages (3 day and 7 day) c) Lightweight aggregates are more effective when samples are pre-damaged at later stage (14 day and 28 day)	[20]

were produced – encapsulated spores, encapsulated nutrients and encapsulated spores and nutrients.

Micro or nano-capsules need to be specially manufactured in laboratories and the encapsulation has to be done along with the manufacture of such capsules. It is important that the capsule and the encapsulated agent remain stable in the concrete matrix. The capsule material should be chosen in a way that the shell is inert to the encapsulated material and highly alkaline concrete matrix. Capsule shell wall thickness is another important parameter which influences the survivability of capsule during concrete mixing. However, shell thickness can be adjusted by changing agitation rate during encapsulation of healing agent and is not much influenced by manufacturing process [22]. Microcapsules of smaller size may be made by varying agitation rate from 200 rpm to 2000 rpm with fine emulsion [22].

Wang et al. [4] used melamine based microcapsules of size 5 μ m which was in the form of microcapsule emulsion in water. Spores with concentration of about 10^9 cells/g of dry weight of microcapsules were encapsulated following a microencapsulation process based on a patented poly-condensation technique [23]. Microcapsules made by this technique have a shell with an outer and inner face which serves different functions. While the inner face is designed to be compatible with the healing agent, the outer face comprises of functional groups which provide good bonding with the applied surface. This is important because capsules are most likely to rupture when the strain is effectively transferred from the matrix to capsules in the event of tensile cracking. The inner face encapsulates liquid containing bacteria. The shell wall may

be made of a wide range of polymer materials like polyurethane, epoxy, polyolefin, silicon resin etc although aminoplast polymers with reactive products of melamine or urea and aldehyde are most preferable [23]. Such materials formed are water insoluble and provide the kind of friability that would be needed when cracks are intercepted.

Glass capsules function well in terms of stability because of its inert nature. However, it is worth noting that after cracking glass may cause alkali silica reaction if there is presence of moisture due to its silica content. Wang et al. [5] used glass capsules to contain bacteria in polyurethane and silica gel. Bio-agents including the bacteria and deposition medium (urea and calcium nitrate) were injected in separate capsules after the capsules were glued together. When the capsules are held together (by glue), it is ensured that both of them will rupture at the same time and the spores will have easy access to the deposition medium for carbonate precipitation. The size and number of the capsule tubes may be adjusted to hold more healing agent depending on the size of member although casting of each such tubes may be difficult in case of in-situ constructions. Bacterial activity was quantified by amount of urea decomposed and calcium carbonate precipitated, also known as ureolytic activity and carbonatogenesis activity respectively. Carbonatogenesis activity is measured by amount of urea decomposed in deposition medium evaluated from conductivity of urea solution. It was noted that higher bacterial activity was observed in case of immobilization in silica gel and also carbonate particles were more evenly distributed compared to those in polyurethane. The attributed reason is difference in viscosity

of polyurethane and silica sol. Silica sol due to its smaller viscosity will result in homogeneous distribution of calcium carbonate. However, higher recovery in strength and permeability was observed in case of immobilization in polyurethane.

3.2. Survival of capsules during concrete mixing

Many studies have used glass capsules to encapsulate healing agent in developing self-healing action [24]. Glass tubes can carry higher amount of healing agent and can be strategically placed at certain locations where cracks are most expected. Another advantage of using glass capsules lie in its brittleness, because it fractures easily when a crack is intercepted. However, the same property makes it prone to premature cracking during concrete mixing unless some form of protection like cement mortar with metallic wire is used [25]. Wang et al. [5] immobilized bacteria in polyurethane and silica gel and placed them inside glass tubes of 40 mm length and 3 mm diameter. In case of silica gel, two glass tubes were used – one with medium and another with bacteria suspension while for polyurethane three tubes were glued. The two component polyurethane was used placed separately in two tubes (one of them with deposition medium) while the third tube accommodated bacteria suspension. To make use of such technique, it must be ensured that both tubes containing polyurethane are ruptured at the same time to enable reaction between the components and aid in healing. The research findings suggested polyurethane with dead and live bacteria performed almost similarly in terms of recovery of permeability. Therefore, it may be understood that polyurethane played an important role in sealing cracks other than being an immobilization medium. Moreover, when tubular capsules are used there may be incomplete release of immobilized bio-agents due to resistive capillary force and negative pressure at both ends. One of the solutions as studied by Li et al. [26] would be controlling crack widths by fibers to increase the capillary attraction of cracks to release the healing agent.

Micro-capsules containing bacteria are more versatile in terms of survivability because they can be dispersed uniformly in concrete easily although some may break due to impact of mixer or hard aggregates. For better survival rate, as proposed by Dry [27], further work is necessary to develop capsules which are flexible during mixing but becomes brittle once concrete hardens. Wang et al. [4] used melamine-based microcapsules of size around 5 μm in emulsion state to encapsulate bacteria spores. Light microscopy was conducted to visualize and compare images of microcapsules before (in water) and after mixing in mortar paste. Comparing images taken before and after mixing, the authors concluded that most of the microcapsules were not broken during mixing. It may be assumed that when bacteria spores or cells are used, survival of bacteria is more important than survival of microcapsules. Rupture of small number of microcapsules during mixing may be acceptable. The reason is if bacteria are released in fresh concrete precipitation of calcium carbonate by the bacterial metabolism may take place which can still contribute to strength [6,28].

Survival of capsules during mixing process also depends on the wall thickness. Thin walled capsules may rupture during mixing while capsules with very thick walls may not rupture or delay the release of the contained bio-agent. For example, Thao [29] found that glass capsules with wall thickness 2 mm were able to release the agent when cracks were intercepted while capsules with 3 mm thickness were too thick to release the agent. This implies that thickness of capsule walls must be accurately designed for efficient self-healing. Dry [30] proposed use of water soluble adhesive to bundle cylindrical capsules together to survive the mixing process. The bundling would increase the net strength of the capsules to resist forces due to mixing or impact of

aggregates; after mixing, the glue dissolves in contact with water and the capsules may be dispersed in the matrix.

3.3. Effect of addition of bio-agents or capsules on concrete properties

Addition of foreign chemicals such as organic nutrients tends to influence mechanical strength of concrete by altering the microstructure [8,31–33]. Besides, capsules form weak links in the matrix and once the contained agent is released capsules act as just voids or holes in concrete. Such voids affect mainly mechanical strength of concrete.

Direct addition of bacteria spores have been found to result in loss in compressive and flexural strength by about 8–10% [14]. Similar finding was reported by Jonkers et al. [8] with spores of *Bacillus cohnii* and *Bacillus pseudofirmus* added to cement stone specimen causing strength reduction of about 10% at 3, 7 and 28 days. Addition of CERUP prepared by Silva et al. [15] to mortar at 0.5% and 1% by weight of cement did not adversely affect compressive strength but higher dosage of 3% and 5% had significant adverse influence on strength.

Experimental results published by Wang et al. [4] showed that addition of nutrients and capsules significantly affect hydration degree, compressive and tensile strength. Addition of 5% microcapsule by weight of cement reduced the compressive strength by up to 34%. Tensile strength was significantly affected with capsule addition above 3%. Addition of nutrients had less profound effect on concrete strength compared to microcapsules. It was noted that calcium nitrate used as precursor enhanced hydration degree and rate of hydration while microcapsules and yeast extract delay hydration. Calcium nitrate in this case may be considered an accelerator additive to concrete. Therefore, the dosage of calcium nitrate must be adjusted carefully. The delay in hydration due to addition of yeast extract was attributed to screening effect of the nutrient that shields the cement particles from reacting with water.

Considerable strength reduction was witnessed by Jonkers [3] when bacteria spores were encapsulated in lightweight aggregates by replacement of granite aggregates. Lightweight aggregates break easily under load and therefore facilitate self-healing by releasing the spores and precursor but such concrete with reduced strength may not be suitable for structural members. Precursors and nutrients for bacteria are added to concrete for self-healing action. It was noted that only calcium lactate when used as mineral precursor do not affect concrete strength but slightly improves it [3,8,14]. Yeast extract and peptone addition reduces compressive strength; especially while peptone is added late strength of concrete may even be lower than early strength [8].

Addition of microcapsules has been reported to reduce water absorption in concrete [4]. Strength reduction is primarily due to alteration of microstructure due to reduced degree of hydration and poor distribution of hydration products caused by addition of nutrients and microcapsules. However, water absorption does not only depend on degree of hydration but also on fraction of open pores in the matrix. Microcapsules added to the matrix may have pore-blocking action while nutrients for bacterial may have water-proofing effect. Thus, it is possible that even if there is strength reduction, durability properties like water absorption may be improved.

Addition of micro-capsules may also affect rheology of concrete which must be studied while developing capsule-based self-healing action in concrete. In urban building sector, concrete very often need to be pumped to considerable height or over long distances and therefore, addition of capsules must not adversely affect concrete rheological properties. Rheology may be affected by capsule material, shape and size. Spherical microcapsules may have lubricating action through reduction of aggregate interlocking

[34]. It reduces plastic viscosity and yield stress and therefore improves flow behavior. However, improvement in rheology would also depend on the capsule shell material. The shell material must not be highly absorptive because absorption of part of mixing water would reduce free water available for slump or flow of fresh paste.

3.4. Sealing ability and recovery of mechanical and durability properties

Recovery of original concrete properties would depend on crack width healed, bonding of the precipitate with the matrix and structure and strength of the precipitate. For effective self-healing, recovery of properties as high as possible to that of original concrete is desired. Healing ability in bio-based self-healing concrete depends on a number of factors including curing condition, local concentration of viable spores and nutrients, age of concrete and healing time. Healing may be achieved in two steps – calcium carbonate precipitate to seal the cracks and reaction of carbon dioxide produced by bacteria metabolism with unreacted portlandite at crack site to form further precipitates. Wide range of crack width has been observed to be healed by the use of bio-based healing agent. Although it depends on a number of factors healing efficiency is best as long as crack width is restricted within 100–200 μm .

3.4.1. Recovery of mechanical properties

Wang et al. [5] encapsulated bacteria cells in PU and silica gel separately within glass tubes. The crack width created to test the recovery of mechanical strength was about 0.35 mm. More precipitation of calcium carbonate was obtained in the case of silica gel, but strength recovery was only about 5%. Specimens containing PU showed strength recovery of between 50% and 80%. However, the action of bacteria in recovering mechanical strength was dubious, because the strength recovery was not significantly different for live and dead bacteria cells. Amount of precipitate was higher in silica gel compared to PU although strength recovery in case of PU was higher. It may be understood that PU, being a good sealing agent, may have contributed mainly to recovery of mechanical strength.

Pei et al. [6] found that addition of bacterial cell wall can increase strength of concrete through three possible mechanisms. Same may be true in case of encapsulated bacteria spores once they become metabolically active. Carbon dioxide produced by bacterial metabolism reacts with unreacted portlandite around the crack face to convert into stronger calcium carbonate. The insoluble calcium carbonate produced by direct precipitation or conversion of portlandite also cause decrease in porosity and increase particle packing efficiency resulting in higher mechanical strength. This is described in particle packing model [35]. Finally, negatively charged bacterial cell may become nucleation sites for cement hydration. However, this is more applicable in young concrete. Although calcium carbonate is a strong component and compatible with cement, recovery of strength may be limited by bonding of calcium carbonate to concrete.

3.4.2. Recovery of durability properties

Efficient self-healing in concrete would mean that the durability and mechanical strength are recovered fully or close to that of the original specimen. Durability is often measured by water permeability and water absorption tests. Healing of cracks would also mean blocking of any void or interconnected pores through, which foreign chemicals may penetrate from air or water. That in turn reduces water permeability and water absorption. Reduction in permeability absolutely by bacterial action is due to pore clogging

by calcium carbonate which is found to have very low solubility [36].

Luo et al. (2015) observed that healing ratio dropped from 83% for 0.1–0.3 mm crack width to 30% when crack width was 0.8 mm. When crack width is high healing products tend to get washed away from the crack face by moisture or any incoming fluid. The drop in healing ratio may be due to loss of repair agent from the crack face or insufficient amount of healing agent to bridge wider cracks. This is also supported by Xu and Yao [14]. Such findings may be true because bacterial metabolism and precipitation may be limited by concentration of bacteria, nutrients and precursor compound at the crack site. The precipitation rate is slow and therefore if the crack width is large precipitates may be washed off by water or other chemicals before the crack is sealed. It was also found that concrete samples submerged in water had higher healing ratio compared to samples subjected to wet curing [13] which may be due to better transport of healing agent due to concentration difference between the matrix and the surface in submerged condition. In the study, highest healing ratio for water curing was observed at early age which means that ample water could penetrate inside and become available for bacterial metabolism. For wet-curing the repair rate was slow and became almost same as water curing at late stage.

Similar observation was made by Wang et al. [4] using microencapsulated spores of *Bacillus Sphaericus* in melamine based capsules. Maximum crack width of 850–970 μm was healed by immersion in water; however highest healed crack area was observed when subjected to wet-dry cycles of water. During the wet cycle moisture could penetrate inside the matrix while during the dry cycle sufficient oxygen was available to the spores. In case of incubation by continuous immersion in water oxygen is not available to the spores and moreover, continuous exposure to water may not be possible in most of the practical cases. Crack area is a better measure of healing than crack width because it considers healing in two directions. Water permeability test showed that concretes with 5% capsules showed lowest final permeability coefficient when exposed to wet and dry cycles. Although the value was close to those with 3% capsules, much less variation in permeability values was observed in specimens with 5%. It may be interesting to observe from the study that concrete with 5% capsules were worse in terms of strength. Therefore, lower permeability may be explained by decrease in porosity by more capsules. This is valid when capsules are intact. After rupture, lower permeability for 5% capsules may be attributable to the waterproofing effect that the contained substance may cause thus causing slightly lower permeability. However, volume fraction of 3% was deemed optimal from the perspective of effect on mechanical strength and reduced water permeability coefficient.

Reduction in water permeability may be significantly influenced by capsule material or immobilization medium used. Wang et al. [5] immobilized bacteria spores in silica gel and polyurethane. In case of immobilization by silica gel, reduction of permeability by two orders of magnitude was observed. Bacterial activity was higher in silica gel and therefore precipitate formed blocked the pores and reduced permeability. However, in case of polyurethane immobilization minimal precipitation was observed and higher reduction in permeability compared to silica gel is attributed to waterproofing effect of polyurethane [5]. Such effect was prominent because PU foam was used to immobilize spores in glass capsules. After rupture the foam could flow out and block the concrete pores. Bacterial action was successful in precipitating calcium carbonate which could only decrease the porosity of the PU foam and had minimal effect on blocking concrete pores directly.

Substantial reduction in permeability, about 68% has been reported by Wang et al. [10] using hydrogel as encapsulation for

Table 2
Review of test methods used to assess healing and recovery of mechanical and durability properties.

Assessment type	Test	Purpose	References	Limitations/challenges
Visualization and determination	Scanning electron microscopy	Visualization of crystal deposited for healing. Rupture of capsules also studied	[4,8,9,13,14,37]	a) Accuracy and visualization is dependent on the location of imaging and resolution used b) Data on uniformity of carbonate depositions may not be obtained from this method
	Infrared analysis	Examination of precipitate	[16]	a) Sensitive to moisture content of concrete b) Minor depositions may be unnoticed because this method captures only prominent depositions [38]
Recovery of durability properties	Environmental scanning electron microscopy	Visualization of mineral precipitate and analysis	[12,16]	Very low pressure need to be maintained which may affect concrete microstructure by dehydration
	Optical microscopy with image analysis	Visualization of crystal deposition and healing rate	[3,9,16]	Visualization depends on resolution of optical microscope which may be limited because of thin sections
	Water permeability (low pressure and high pressure)	Determination of permeability coefficient post healing give idea on the flow of water through healed cracks	[3–5,9,10]	a) Measurement of permeability is sensitive to composition of concrete, crack size developed and orientation of cracks b) Air and water tight seal is required for accurate measurement
Recovery of mechanical properties	Water absorption	Measurement of susceptibility of healed concrete to water penetration under capillary force	[4,9]	c) Accuracy would also depend on sample preparation and finishing
	Bending test (3-point and 4-point)	Measure recovery in strength due to self-healing	[9,14]	a) UPV results are sensitive to certain parameters including age of specimen to be tested, water-cement ratio, curing condition and compaction. Reliability of results would depend on control of these parameters
	Ultrasonic pulse velocity (UPV)	Measurement of mechanical properties of concrete during damage and healing	[14]	b) Concrete specimen should be sufficiently dried because presence of voids filled with water may produce misleading UPV results

bacteria spores and bio-reagents. Maximum crack width of 0.5 mm was healed although there was high variation of healing ratio (40–90%) even for 0.3–0.4 mm cracks. However, it is an improvement compared to when only hydrogel was used. Better healing may be expected due to proportionate distribution of spores and bio-reagents when they are encapsulated together in hydrogel. Due to this encapsulation strategy, in the event of cracking bacteria would have ready access to the nutrients and precursor compound. Moisture absorption and retention property of hydrogel also helped in higher bacterial action. Moreover, reduction in permeability may also be boosted by some autogenous healing facilitated through internal curing by hydrogel in addition to bacterial precipitation.

Water absorption results obtained by Wang et al. [9] showed substantial improvement when bacteria immobilized in diatomaceous earth was used. Water absorption was reduced by one-third and 50% when cracked specimens were incubated in deposition medium (containing yeast extract, calcium nitrate and urea) and water respectively. During the process of healing calcium ion and urea from deposition medium could penetrate inside and boost precipitation of calcium carbonate. This effect was more prominent probably because the concrete specimens were only 14 day old when cracks were introduced and therefore, nutrients and calcium from the deposition medium could penetrate inside the concrete matrix easily. However, it is also stated by Wang et al. [9] that high calcium ion concentration reduces amount of hydroxyl ion from dissolution of calcium hydroxide causing drop in pH. This may slow down precipitation because of drop in optimal alkalinity for bacterial action. In case of older concrete where dense microstructure is formed due to more complete hydration, calcium ions cannot penetrate inside the concrete easily when immersed in deposition medium. Therefore, the calcium concentration would be high only at the surface restricting the bacteria spore from accessing the external supply of calcium and deposition source.

A summary of techniques and tests to test crack sealing and recovery of mechanical and durability properties after self-healing has occurred has been presented in Table 2.

The outcome of this review can be used to evaluate robustness of capsule based self-healing strategies for sustainable infrastructure in terms of the six highlighted robustness factors proposed by Li and Herbert [39]– shelf life, pervasiveness, quality, reliability, versatility and repeatability. Robustness of two techniques – direct addition of bacteria and reagents to concrete and addition of bacteria via encapsulation are summarized in Table 3 in light of these six robustness factors for evaluation of efficiency of capsule based self-healing.

4. Conclusion and scope of future work

It is a fact that the initial cost of incorporating bio-agents may be high at present. There may be two possible ways to make it commercially viable – by reducing the cost of production of bio-agents or by designing the self-healing action for longer life so that it works well under multiple cycles of loading and adverse environment condition to which structures are often subjected to. Presently, high cost is incurred due to a number of repair activities to handle several deterioration events throughout an infrastructure's life. This is explained by life-cycle cost model conceptualized by van Breugel [40]. Repair cost may in some cases exceed the initial construction cost. On the contrary, if infrastructures are built with self-healing concrete that is designed to function under multiple damages, very minimal cost may be accumulated over the lifetime although initial cost may be higher than normal concrete.

Table 3
Summary of efficiency of bio-based self-healing in light of six robustness factors.

	Shelf life	Pervasiveness	Quality	Reliability	Versatility	Repeatability
Direct addition of bio-agents in concrete	Bacteria spores are more stable than live cells but harsh environment of concrete reduce lifetime of spores to only 1–2 months [3]	Uniform dispersion of bio-agents including nutrients is possible [12,13]	Sealing ability depends on crack width. Crack width up to 450 μm has been sealed [15] Inadequate data to comment on recovery of strength although flexural strength recovery of about 25% recorded [14]	Reliable sealing ability and recovery of concrete properties on short term. Long term performance not studied	Highly dependent on crack width, cracking age and presence of moisture and nutrients [13]	Inadequate results to establish healing for multiple loading cycles
Addition of encapsulated bio-agents	Survival period and rate of bacteria is enhanced due to encapsulation. Viability up to 6 months has been studied [3]	Uniform distribution of capsules or immobilization medium containing bacteria possible [3,4]	Sealing ability depends on crack width. Maximum crack width of 970 μm healed [3,4] a) High recovery of durability measured by water permeability or water absorption tests [3,4,9] b) Inadequate data to comment on recovery of strength	Reliable sealing ability and recovery of concrete properties on short term. Long term performance not studied	Effective healing when moisture is present [3,4,10] and crack width is restricted	Inadequate results to establish healing for multiple loading cycles

Design of capsules is an integral part of self-healing. More research should be focused on development of capsules that least affect concrete properties. Moreover, there is very little evidence of bio-based self-healing performance under fatigue. Fatigue performance of bio-based self-healing concrete would depend on release behavior of capsules. Healing under multiple loading cycles may be achieved through controlled release or 'smart release' from capsules. Dong et al. [41] worked on development of smart releasing microcapsules targeted at healing concrete degradations, especially corrosion of reinforcement bars triggered by reduced alkalinity of concrete matrix. However, it is not very established at this point of time and more research is needed on application of such smart capsules in bio-based self-healing. Capsules at nano-scale may be explored to reduce the size of weak spots created in mortar due to introduction of capsules. Urea formaldehyde (UF) Capsules with diameters 220 nm and thickness of 77 nm with more uniform shell wall were achieved successfully by a sonification technique although only chemical healing agent has been encapsulated so far [42]. If the capsule material can be designed to be compatible with bio-agents, its application may be extended to bio-based self-healing concrete. However, it must be ensured that the nano-particle debris do not agglomerate in the concrete matrix because nano-particle agglomeration sites may be spots for crack initiation in the matrix.

Findings, so far, suggest that crack closure time for bio-based healing with suitable curing conditions takes long time which typically span at least between two to three weeks. This is due to lower precipitation rate by bacteria in concrete environment. More interdisciplinary research may be needed to produce genetically modified bacteria culture which may thrive longer and precipitate at faster rate. If such research becomes successful it may be hoped that in the future cracks of bigger widths may also be healed in shorter time. Control of crack width is also an important factor to achieve faster and efficient healing by biological action [21]. Use of hybrid fiber could be explored as a way to control crack width, that can offer high recovery of original properties after healing [43,44]. Combination of steel and polymer fibers not only restrict crack width, but also provide an anchor for healing products to attach near the crack face [43].

Further research is needed on the efficacy of self-healing in actual site environment. These studies should aim to enhance the service life, reduce cost and look at the environmental and social benefits in detail. It will be useful to study how using bio-based self-healing in concrete contribute to climate change adaptation [37,45]. Sustainability assessment methods, such as life cycle assessment, can also be applied to improve the life cycle of bio-based self-healing systems [46–48]. In this regard, bio-based healing agents may be combined with carbon sequestering material [49] to develop more sustainable and 'green' self-healing composites. Finally, specifications and test standards should be developed to assess bio-based self-healing performance in buildings.

Given the research focus on bio-based self-healing materials in past one decade, its implementation in building sector in near future may be expected. However, the current technological and technical barriers must be first addressed to make it fit for wide-spread industry practice.

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