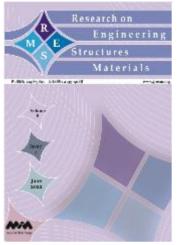


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Review Article

Self-healing concrete techniques and performance: A review

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Abstract

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Keywords:

Calcium carbonate; Self-healing; Concrete cracks; Autogenous selfhealing; Autonomous selfhealing Modern society faces the duality of rapidly expanding structure, making concrete one of the world's most traded materials. However, cement manufacturing can pollute the environment by releasing approximately one tonne of CO₂ for every tonne of cement produced. Concrete cracks can provide superior access for aggressive substances such as chlorides and sulfates, resulting in structural deterioration. So, to fix concrete cracks, different traditional methods were used, which use cement and some chemical agents that are hazardous to the environment. Because of the environmental issues and sustainability challenges associated with cement and concrete, it is preferable to reduce the amount of cement used by developing promising and unique solutions to enable quick crack healing in concrete and extend the structure's lifetime. Therefore, incorporating self-healing mechanisms into construction materials has been proposed to improve their performance and durability while reducing the need for maintenance and repair. This review assesses the performance and causes of autogenous and autonomous self-healing techniques. The autogenous technique occurs naturally due to inherent material properties, while the autonomous technique uses various healing agents, such as chemical or biological substances. Both techniques rely on forming calcium carbonate (CaCO₃) crystals as the principal agent for concrete healing. Previous findings showed that the autogenous technique has limited efficacy in repairing larger cracks with a width exceeding 0.3mm. In contrast, autonomous techniques have shown successful repai of cracks exceeding 2mm in width. The application of an autonomous methodology in the field of concrete has resulted in significant results, such as effectively repairing large cracks, enhancing structural integrity, and substantially decreasing permeability levels from high to exceedingly low levels.

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1. Introduction

Concrete is a highly prevalent construction material worldwide, with an estimated annual production of approximately 6 million cubic meters [1]. While using concrete as a final product does not harm the environment, it is essential to note that producing concrete components has adverse effects. Specifically, the global annual consumption of Portland cement. Concrete often exhibits cracks due to excessive tensile stresses or environmental conditions [2, 3]. Accordingly, rehabilitation and maintenance of concrete structures are essential. For example, the direct cost of maintaining and repairing concrete bridges in the United States was nearly \$4 billion annually [4-7]. Repairing and rehabilitating existing structures account for 50% of annual construction costs in Europe [8] and 50% in the United Kingdom [6, 9, 10].

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Traditional methods, including sealing, routing, stitching, grouting, chemical injection, and carbon fiber reinforcement, have been extensively used over time. Nevertheless, these methods possess distinct disadvantages as they can be time-consuming, mainly when dealing with larger or more complex cracks. Moreover, the conventional crack healing processes may exhibit inconsistencies, which can be attributed to many factors, such as labor skill, material quality, and environmental conditions. Also, these methods often repair large cracks but are inappropriate for deep and small cracks [11]. Repairing agents can be hazardous to the environment and expensive [10]. Furthermore, the utilization of cement is required in the conventional procedure of concrete crack healing, which leads to an increase in carbon dioxide (CO_2) footprint [12]. Finally, traditional methods might require continuous maintenance and periodic reapplication of healing agents to maintain long-term efficacy.

Presently, self-healing concrete is regarded as a durable repair technique that has captured the interest of researchers [13, 14]. Self-healing concrete exhibits properties similar to the human body, whereby injuries and wounds can autonomously heal without external intervention [15]. Over the past decade, there has been a notable expansion in research related to self-healing cementitious composites. This paper aims to provide a comprehensive overview of various self-healing techniques and their respective performance. The concept of self-healing concrete, covering both autonomous and autogenous healing, was initially proposed. This review offers an in-depth review of the mechanisms by which this material achieves crack healing and the specific steps involved in the healing process. Also, the factors affecting both techniques were reviewed.

2. Self-Healing Techniques

Autogenous and autonomous are two prevalent self-healing techniques extensively employed for concrete healing. The autogenous technique aims to improve the natural mechanism of crack healing. Every concrete structure has a limited autogenous healing potential [16]. This technique includes only the material's original components due to their specific chemical structure and promotes healing under environmentally favorable systems [7]. In addition, owing to its inherent characteristics, concrete possesses microreservoirs containing sparsely distributed un-hydrated cement particles, which facilitate the process of self-repair [17]. The autogenous self-healing technique was initially observed about 200 years ago, in 1836 by the French Academy of Sciences [17-24]. Since the early 19th century, researchers have explored the effectiveness of this method for repairing cracks in pipes, culverts, and other water infrastructure [25-27].

On the other hand, the autonomous technique aims to modify concrete by adding various healing agents to the concrete matrix so that the crack heals autonomously after its formation [7]. In this approach, the repairing agents are pre-buried in the concrete mixture and can automatically heal cracks as they occur, saving potential costs compared to traditional methods [28].

The primary mechanism employed by both techniques to facilitate the healing of cracks involves the deposition of white crystalline calcium carbonate ($CaCO_3$) material onto the surface of the crack. This material can be generated in natural (autogenous) or artificial (autonomous) ways. In addition, $CaCO_3$ possesses a rough crystal morphology that exhibits a high propensity for surface adhesion and self-sustaining growth [29]. The subsequent sections provide a comprehensive analysis of both techniques.

3. Autogenous Self-Healing Technique

Concrete autogenous self-healing is assumed to be related to several mechanisms [30]. As per RILEM [31], autogenous self-healing can be attributed to three primary physical, chemical, or mechanical factors, as illustrated in (Fig. 1) [7, 32].

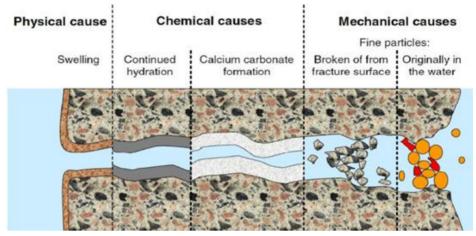


Fig. 1. Different causes of autogenous self-healing [7]

3.1. Causes of Autogenous Self-Healing Technique

3.1.1. Physical Cause

Physical cause includes the hydrated cement paste (HCP) enlargement next to the crack opening [19]. This phenomenon arises as HCP absorbs water, which goes through the space between the components of HCP, resulting in an expansion of calcium-silicate hydrate (C-S-H) gel [33]. It can be described as the volume increase due to the matrix's saturation [34].

3.1.2. Chemical Causes

The chemical causes can be divided into two mechanisms: continued hydration and calcium carbonate formation (carbonation) [18, 31].

· Continued hydration

Despite the potential impact of continued hydration on self-healing, it has not gained as much attention as the carbonation process [35]. The by-products of the continued hydration process can have twice the volume of cement [7, 36-38], filling spaces previously filled with water and causing crack healing [34]. However, if the crack width is narrow (less than 0.1 mm) and the swelling and hydration happen simultaneously, the crack may close independently. The effect decreases as the cracks get wider [7, 34].

Carbonation

According to previous research, the carbonation effect on autogenous healing is more crucial than that of continued hydration [17, 31, 33, 36]. First, this process happens in concrete due to dissolved soluble composites moving out of the concrete matrix. Following this, the formation of $CaCO_3$ begins due to the interaction of calcium ions derived from portlandite (Ca(OH)2) with carbon dioxide molecules encased in water, as shown in Eqs. (1-2) [1, 25, 39]. Finally, $CaCO_3$ appears as white crystals deposited on the surfaces of cracks.

$$CaO + H_2O \rightarrow Ca(OH)_2 \tag{1}$$

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3 + H_2O \tag{2}$$

Also, $CaCO_3$ can be formulated according to the following reactions, as shown in Eqs. (3-4) [40], depending on the reactant's temperature, concentration, and pH [7, 41, 42]. Calcium ions (Ca^{2+}) that exist in the concrete components react with carbonates (CO_3^{2-}) or bicarbonates (HCO_3^{-}) available in water, creating insoluble $CaCO_3$, which causes the healing of cracks [36, 40, 41].

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3$$
 $pH water > 8$ (3)

$$Ca^{2+} + HCO_3^- \rightarrow CaCO_3 + H^+ \qquad 7 < pH \text{ water} < 8$$
 (4)

3.1.3. Mechanical Cause

This technique, also known as the self-tightening technique, does not involve any chemical reaction. Instead, it involves the obstruction of the crack by tiny particles at the crack face or the passage of small impurities through the crack [7, 32, 33], as shown in (Fig. 1).

3.2. Factors Affecting Autogenous Self-Healing Technique

Numerous parameters may affect the autogenous rate of healing, which are discussed in detail in the following sections.

3.2.1. Water/Cement Ratio

Enhancement of autogenous healing can be achieved through improving continuing hydration by increasing cement content or reducing the W/C ratio [26, 34, 43]. Theoretically, a W/C ratio of 0.22 is adequate for achieving complete hydration. However, according to sources [17, 26], approximately 30% of the cement particles present in conventional concrete have yet to undergo the hydration process. Even so, the hydrated cement particles may still include a small, slowly-hydrating, un-hydrated core, which will hydrate later [34, 44]. When unhydrated particles are contacted with external water after cracking, they hydrate, healing cracks and voids. Better and more efficient mixing processes result in fewer unhydrated cement particles reacting with air moisture [19].

3.2.2. Cement and Additives Type

Based on continued hydration, self-healing can be affected by the cement particle size [34]. The clinker content controls the amount of Ca^{2+} ions and, as a result, the matrix's capability to develop $CaCO_3$ precipitation products [45]. The influence of particle size and coarse cement mixing content on autogenous healing was investigated using ultrasonic measurements. The self-healing ratio increased with cement particle diameter for equal mixing content of coarse cement [45].

Different additives can be included in the concrete mixture, affecting the healing rate. For instance, incorporating fibers can enhance the healing processes due to their participation in reducing crack width. However, it affects workability when adding a large amount of fibers [19]. A uniformly distributed sufficient fiber content can enhance mechanical properties and tighten the crack [46, 47].

Fly ash, silica fume, and blast furnace slag are known as aluminosilicate materials, which can improve autogenous self-healing ability through a pozzolanic reaction. In alkaline conditions, silicic acid (H_4SiO_4/SH) can be formed from silicate species dissolved in pozzolanic material. Silicic acid can react with dissolved $Ca(OH)_2$ to produce C-S-H and

water [7]. These reactions can give a level of self-healing capacity due to the formation of C-S-H, according to Eqs (5-6) [7].

$$3Ca(OH)_2 + 2H_4SiO_4 \rightarrow 3Ca_2 + 2H_2SiO_4^{2-} + 4H_2O + 2OH^{-}$$
 (5)

$$3Ca^{2+} + 2H_2SiO_4^{2-} + 4H_2O + 2OH^- \rightarrow 3CaO . 2SiO_2 . 7H_2O$$
 (6)

Like the C-S-H formed during the hydration of un-hydrated cement particles, the C-S-H formed during the pozzolanic process can repair small cracks. Since the pozzolanic reaction is pH-dependent, it takes much longer than cement hydration [7].

3.2.3. Water Availability During The Healing Stage

Water availability is crucial for healing since it is necessary for chemical reactions to promote $Ca(OH)_2$ dissolution from the concrete matrix next to the crack surface [7, 34, 48-50]. Several researchers found that the specimens healed through water immersion performed better than those cured through humidity chambers or air curing [34, 51]. Few investigations revealed that the self-healing phenomenon was more present in wet and dry cycles than in completely submerged conditions because of the abundance of CO_2 in the atmosphere [49, 52].

3.2.4. Concrete Age

Concrete age is vital to autogenous healing [34]. Early-aged concrete has a better healing ability than old ones due to un-hydrated cement in early-age concrete [7, 53, 54]. Also, early-age concrete can develop new C-S-H gels, which can be continued by combining the two chemical processes mentioned above. Leading to primarily $CaCO_3$ deposits for crack closure at later ages [49]. It was realized that healing efficiency diminishes with the sample's age [48, 55]. In contrast, self-healing at later ages is attributed to the formation of calcium carbonate [48].

3.2.5. Crack Width

The dimensions and patterns of cracks can influence the healing ability of concrete. [49]. By reducing the crack's width and improving the healing mechanism, larger cracks can be repaired more rapidly and efficiently. This means that the autogenous intrinsic healing potential of cement-based composites can enhanced by restricting and managing the crack width [49]. For example, for the crack width, in the first 24 hours, cracks with an effective width of less than 50 microns were reduced to 20 microns; in the following seven days, cracks with an effective width of 50 to 100 microns shrank to 20 microns. [17, 45, 55].

As mentioned in previous studies, the autogenous self-healing technique can only heal small cracks of 0.1-0.2 mm [16, 26, 31, 36, 43, 56]. However, other researchers mentioned that this technique could heal cracks up to 0.3mm in width [23, 31, 51, 57]. A high crackhealing potential of concrete structures is advantageous because it enhances the material's durability [39]. Thus, an alternative self-healing mechanism will likely be incorporated to improve the durability of such relatively cheap and environmentally sustainable concrete [25]. Moreover, the current policy seeks to reduce the cement required in a concrete mixture because its production is environmentally unfriendly due to high energy consumption and CO₂ emissions [58, 59].

4. Autonomous Self-Healing Technique

Autonomous self-healing concrete involves the incorporation of a suitable healing agent, either chemical or biological, into the concrete during its production, thereby facilitating the healing process. (Fig. 2) offers additional clarification regarding the diverse techniques employed in the process of biomineralization.

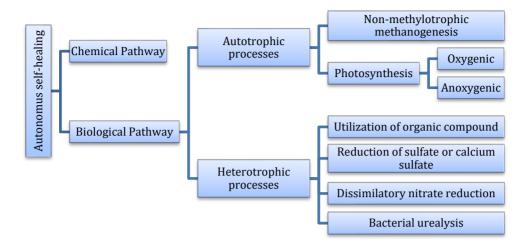


Fig. 2. Autonomous pathway processes.

4.1. Chemical Self-Healing

Chemical self-healing is an autonomous healing technique carried out by adding chemical agents to the concrete matrix during the mixing. Glue, calcium sulfoaluminate, and crystalline admixtures are the most commonly used chemical agents [34, 60]. The main drawback of this process is the possibility of achieving the healing ability before a crack formation [34]. Different scientists examined chemical agents like:

• Aqueous sodium silicate (Na₂SiO₃) reacts with Ca(OH)₂ from the concrete, forming C-S-H as a result, as shown in Eq. (7) [60].

$$Na_2SiO_3 + Ca(OH)_2 \rightarrow (CaO.SiO_2).H_2O + Na_2O$$
 (7)

Crystalline admixture (CA) is a distinct category of permeability-reducing admixture (PRA) [34]. Crystalline admixture reacts with tricalcium silicate (3Ca0 – SiO₂) to produce modified C-S-H and a precipitate made of calcium and water molecules as a by-product of their reaction, as shown in Eq. (8).

$$3CaO - SiO_2 + M_x R_x + H_2 O \rightarrow Ca_x Si_x O_x R - (H_2 O)_x + M_x CaR_x - (H_2 O)_x$$
(8)

Where $3CaO - SiO_2$ is tricalcium silicate, M_xR_x is the crystalline promoter, $Ca_xSi_xO_xR - (H_2O)_x$ is the modified C-S-H + pore-blocking precipitate.

• Shrinkage compensators of calcium sulfoaluminates were used as self-healing agents [61, 62]. Since their expansive reaction has the potential to fill cracks, they have the potential to be self-healing agents. Compared to other mineral additions and OPC, the impact of shrinkage compensators can be visually inspected through the closure of early-age cracks of size 0.1–0.3 mm.

The best results for self-healing were obtained by combining two different admixtures, shrinkage compensators and crystalline admixtures [62]. However, it was shown that expansive admixtures respond best to adding a single mineral due to the crystals' tendency to overgrow [63].

4.2. Biological Self-Healing

In this technique, micro-organisms were chosen as healers due to their adaptability in healing cracks as a green alternative to traditional methods. This approach is known as "bacterial concrete" [4, 11, 39, 43, 64, 65]. Bacterial concrete has recently gained popularity due to its superior healing capacity, which should last for the structure's lifetime [17, 34]. The bacterial concrete technique is based on mineral-producing bacteria that deliver good results when sprayed, applied into the crack, or added to the concrete matrix [42, 65].

Bacteria make long-lasting healing possible, and this process is known as microbiologically induced calcium carbonate precipitation (MICP). MICP acts as a binder, filling cracks and binding the concrete constituents. Bio-mineralization is the basic mechanism of incorporating bacteria in concrete using different processes to produce CaCO₃ [56]. Generally, biomineralization can be divided into two significant processes: heterotrophic and autotrophic. Heterotrophic processes precipitate more CaCO₃ than autotrophic ones [66].

4.2.1. Autotrophic Processes

The autotrophic term refers to the organisms that produce complex organic compounds, such as carbohydrates, with the aid of light energy, as in photosynthesis, or chemical reactions, as in chemosynthesis. The following sections list autotrophic pathways.

• Non-methylotrophic methanogenesis

Non-methylotrophic methanogenesis is a set of micro-organisms called methanogens. This approach is more commonly observed in marine sediments. Examples of methanogens micro-organisms are those of the Methanobacterium species [67]. This type of bacteria provides methane (CH_4) metabolically under anaerobic conditions [67, 68]. In this process, methanogenic archaebacteria use carbon dioxide (CO_2) and hydrogen (H_2) in strict anaerobiosis to make methane (CH_4), as shown in Eq. (9).

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O$$
 (9)

After that, CH₄ is converted into bicarbonate through anaerobic oxidation with the assistance of sulfate (SO_4^{2-}), which functions as an electron acceptor, as shown in Eq. (10). Then, the produced bicarbonate (HCO_3^-) reacts with calcium ions (Ca^{2+}) resulting in $CaCO_3$ formation, as shown in Eq. (11).

$$CH_4 + SO_4^{2-} \to HCO_3^- + HS^- + H_2O$$
 (10)

$$Ca^{2+} + HCO_3^- \to CaCO_3 + CO_2 + H_2O$$
 (11)

• Photosynthesis process

Photosynthetic bacteria can heal cracks through oxygenic and anoxygenic photosynthesis. In general, cyanobacteria and microalgae micro-organisms tend to display more significant activity levels in aquatic environments [69]. Different kinds of electron donors are required for oxygenic (cyanobacteria) and anoxygenic (purple bacteria) photosynthesizing organisms to produce methanal (CH_2O) successfully [68]. In the oxygenic photosynthesis, water donates electrons to produce oxygen, as shown in Eq. (12). But in anoxygenic photosynthesis, hydrogen sulfide H_2S acts as electron donor, therfore oxygen is not produced as shown in Eq. (13) [68, 70].

$$CO_2 + H_2O \xrightarrow{Oxygenic\ photosynthesis} CH_2O + O_2$$
 (12)

$$CO_2 + 2H_2S + H_2O \xrightarrow{Anoxygenic\ photosynthesis} CH_2O + 2S$$

$$+ 2H_2O$$
(13)

The remaining reactions are the same in both groups. CO2 is taken out of bicarbonate solutions, as shown in Eq. (14), to make carbonate compounds. This causes a concentrated rise in pH, which, as shown in Eq. (15), speeds up the creation of $CaCO_3$ when calcium ions are present, as shown in Eq. (16) [66].

$$2HCO_3^- \leftrightarrow CO_2 + CO_3^{2-} + H_2O \tag{14}$$

$$CO_3^{2-} + H_2O \leftrightarrow HCO_3^{-} + OH^{-}$$
 (15)

$$Ca^{2+} + HCO_2^- + OH^- \to CaCO_3 + H_2O$$
 (16)

4.2.2. Heterotrophic Processes

The term heterotrophic means that micro-organisms must acquire materials and energy from an alternative source to create a substance [66]. Also, they need organic carbon to grow [56]. This process can be achieved using different methods discussed in the following sections.

• Utilization of organic compounds

Bacteria with a calcium source represent a two-component healing agent in this process. Different types of bacteria, including Anthrobacter, Rhodococcus, and Bacillus, are necessary due to their ability to facilitate the metabolic conversion of organic molecules containing calcium, such as calcium lactate, calcium acetate, or calcium formate, into $CaCO_3$ [56, 66].

The following equations show the metabolic conversion of some organic compounds in the presence of bacteria. Calcium lactate metabolic conversion is shown in Eq. (17) [10, 64]. The metabolic conversion of calcium-formate with portlandite exists in the paste matrix shown in Eq. (18) [25]. Also, calcium acetate's metabolic conversion is shown in Eqs. (19)-(20) [67]. Eventually, bacteria act as a catalyst in this process. In addition to $CaCO_3$ formation, bacteria can produce CO_2 , which is trapped inside the concrete and enters a reaction similar to the carbonation process but with faster rates, as described in Eq. (2). This improves the concrete standard carbonation reaction [43].

$$CaC_6H_{10}O_6 + 6O_2 \xrightarrow{Bacteria} C_aCO_3 + 5CO_2 + 5H_2O$$
 (17)

$$Ca(CHO_2)_2 + Ca(OH)_2 + O_2 \xrightarrow{Bacteria} 2C_aCO_3 + 2H_2O$$
 (18)

$$CH_3COO^- + 2O_2 \xrightarrow{Bacteria} 2CO_2 + H_2O + O^-$$
 (19)

$$Ca^{2+} + 2HCO_3^- + O_2 \xrightarrow{Bacteria} C_aCO_3 + H_2O + CO_2$$
 (20)

• Reduction of sulfate or calcium sulfate

In this process, the healing usually occurs under anoxic conditions using sulfate-reducing bacteria (SRB) such as (anaerobic, prokaryotes, and morphologically) [70]. The abiotic dissolution of gypsum ($CaSO_4$. H_2O) provides a media which is reach in both sulfate (SO_4^{2-}) and calcium ions (Ca^{2+}) as shown in Eq. (21). In the existence of organic matter with no oxygen, SRB can diminish sulfate (SO_4^{2-}) to hydrogen sulfide (H_2S) and release bicarbonates (HCO_3^{-}) as shown in Eq. (22). After that, hydrogen sulphide degasses, increase the pH level leading to calcium carbonate precipitation as shown in Eq. (23) [66, 69, 70].

$$CaSO_4. H_2O \xrightarrow{Abiotic \ dissolution} Ca^{2+} + SO_4^{2-} + 2H_2O$$
 (21)

$$2(CH_2O) + SO_4^{2-} \to HS^- + HCO_3^- + CO_2 + H_2O$$
 (22)

$$Ca^{2+} + HCO_3^- + OH^- \to CaCO_3 + H_2O$$
 (23)

Conflicting points of view were discovered regarding utilizing the SRB strain of bacteria as a healing agent in cementitious materials. The utilization of SRB has been noted to have deleterious effects on concrete, as reported in a previous study [67]. However, other studies revealed that sulfate-reducing bacteria in concrete at different concentrations significantly improved the material's compressive, tensile, and flexural strengths. As per the findings, the utilization of this pathway is not common owing to elevated levels of sulfate [71].

• Dissimilatory nitrate reduction (Denitrification)

Calcium carbonate can be efficiently produced by utilizing nitrate-reducing bacteria, which facilitate the generation of nitrogen and carbon dioxide during the process. This process can be defined as a respiratory process that reduces nitrate to nitrite, nitric oxide, nitrous oxide, and nitrogen gas. As a result, nitrogen, carbon dioxide, and water are released in this process, as shown in Eq. (24). In addition, hydrogen ions (H+) consumption during the denitrification process increases pH levels, leading to carbonate formation, as shown in Eq. (25). Finally, calcium ions combine with carbonate to generate calcium carbonate, as shown in Eq. (26) [67].

$$NO_3^- + H^+ \xrightarrow{Denitrification} CO_2 + N_2 + H_2O$$
 (24)

$$CO_2 + 2OH^- \rightarrow CO_3^{2-} + H_2O$$
 (25)

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3 \tag{26}$$

The effects of nitrate-reducing bacteria on the mechanical properties of concrete have been examined through an investigation of Diaphorobacter nitroreducens, a strain of bacteria that exhibits denitrification bacterial activity. The research findings indicated a rise in compressive strength, as evidenced by the data reported [66, 72]. The pathway in question is primarily associated with Denitobacilus, Thiobacillus, Alcaligenes, Pseudomonas, Spirilium, Achromobacteri, and Micrococcus bacterial species. Nevertheless, this methodology has not undergone comprehensive investigation, and its applicability is limited.

• Bacterial urealysis or (urea hydrolysis)

Several bacteria crucial for the environment and medicine can synthesize the enzyme urease (urea amidohydrolase) [73]. The application of urea hydrolysis as a healing agent in the autonomous self-healing mechanism of concrete was first introduced in 1995 [74]. This strain of bacteria facilitates calcite formation via a complex sequence of biochemical reactions. This bacteria is widespread because it can hydrolyze urea quickly and generate carbonate ions without using unnecessary protons [66]. According to research findings, the bacterial urease enzyme-mediated decomposition of urea is the most commonly employed healing technique in engineering applications [56, 75]. Calcium carbonate precipitation in this process is mainly affected by the amount of the enzyme urease formed by the bacteria [75].

The first step in the reaction in the presence of the bacteria catalyzes the hydrolysis of urea $(CO(NH_2)_2)$, leading to the production of carbamic acid (NH_2COOH) and ammonia (NH_3) , as shown in Eq. (27) [73].

$$CO(NH_2)_2 + H_2O \xrightarrow{Bacteria} NH_2COOH + NH_3$$
 (27)

On further hydrolysis, carbamic acid spontaneously decomposes into ammonia and carbonic acid (H_2CO_3), shown in Eq. (28) [70].

$$NH_2COOH + H_2O \rightarrow NH_3 + H_2CO_3$$
 (28)

Carbonic acid subsequently equilibrates in water, forming bicarbonate (HCO_3^-) , ammonium (NH_4^+) , and hydroxide ions (OH^-) as shown in Eqs. (29)-(30).

$$H_2CO_3 \to HCO_3^- + H^+$$
 (29)

$$2NH_3 + 2H_2O \to 2NH_4^+ + 2OH^- \tag{30}$$

This reaction raises the pH level and produces carbonate ions by shifting the bicarbonate equilibrium, as shown in Eq. (31).

$$HCO_3^- + H^+ + 2NH_4^+ + 2OH^- \rightarrow CO_3^{2-} + 2NH_4^+ + 2H_2O$$
 (31)

Because of their negative charge, bacterial cell walls attract cations from their surroundings, including calcium ions, which the bacteria then deposit on their surface. Nucleation of calcium carbonate crystals occurs at the cell surface as a result of Ca^{2+} reactions with carbonates CO_3^{2-} , as depicted in (Fig. 3). Finally, as indicated by Eqs. (32)-(33), an increase in carbonate concentration causes a rise in supersaturation, precipitating CaCO₃ around the cell in the presence of soluble calcium ions.

$$Ca^{2+} + Cell \rightarrow Cell - Ca^{2+} \tag{32}$$

$$Cell - Ca^{2+} + CO_3^{2-} \rightarrow Cell - CaCO_3^{-}$$
(33)

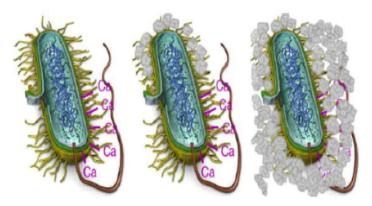


Fig. 3. CaCO₃ precipitation on bacteria cell wall [56]

The hydrolysis of urea has numerous potential benefits compared to the carbonate-generating approach. The hydrolysis of urea can be simply regulated, generating superior quantities of carbonate in a short period [20]. In contrast to the urease hydrolysis mechanism, the metabolic conversion of calcium lactate does not produce massive amounts of ammonia, drastically increasing the risk of reinforcement corrosion [76].

4.2.3. Factors Affecting Biological Concrete

(Fig. 4) clarifies the factors upon which the efficacy of bacterial self-healing concrete depends, which will be explained in subsequent sections.

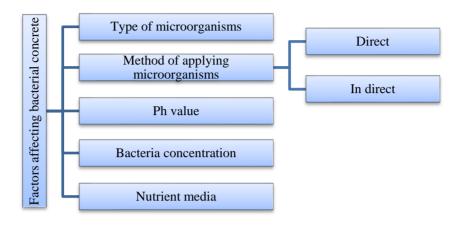


Fig. 4. Factors affecting bacterial concrete

• Type of micro-organisms

Different types of bacteria can be used in concrete, such as anaerobic and aerobic bacteria. For aerobic bacteria, using Bacillus subtilis improved compressive strength [1, 6, 10, 26, 77-84]. Using Bacillus pasteruii improved the strength and permeability of concrete [29, 85-90]. Bacillus chonii does not affect mechanical properties [25, 28, 64, 91]. Bacillus pseudofirmus did not affect compressive strength [91]. However, it was mentioned in other research that using this type of bacteria reduced compressive strength [25, 92]. Anaerobic bacteria such as E. coli did not affect strength, but Shewanella improved compressive strength [93].

• Methods of incorporating bacteria

Bacteria can be incorporated into the concrete matrix using various methods, broadly classified into direct and indirect.

In the direct method, bacteria are directly incorporated into the concrete matrix through the water of the mixing process. Unfortunately, this method reduced bacteria viability due to bacteria crushing during mixing and squeezing after concrete hardening [64, 94]. When spores were used, survivability improved, but it also changed the porosity during maturation, resulting in decreased viability and survival shortened to 1-2 months [64] or four months [64, 95]. Almost one-half of the bacterial cells were kept in a vegetative state in the matrix after 330 days with proper bacteria and nutrients [96].

The indirect method can be classified into two methods. The first method is the adsorbed method. In this method, different immobilizers were used to carry the bacteria to preserve it from the harsh environment of the concrete [97]. The immobilization technique has been reported to be effective in maintaining an efficient mineral formation capacity to incorporate bacteria in self-healing concrete over time [28]. The material used as a bacterial immobilizer must be able to immobilize bacterial spores effectively. Several trials have been conducted to develop porous bacteria immobilizers [39, 98, 99]. When the absorption factor of the immobilizer increases, the healing efficiency increases since it can

ensure adequate oxygen, water, and a growing area for bacteria after the formation of the crack. Immobilizers not only help in the survival of bacteria, but some also impact the mechanical characteristics of concrete [10]. Different immobilizers were utilized in self-healing concrete, such as iron oxide nano/microparticles and bentonite nano/microparticles [97], lightweight aggregate (LWA) and graphite nanoplatelets (GNP) [10], zeolite powder [100], and expanded clay particles [64]. Different recycled materials were used, such as recycled brick aggregate [83], recycled concrete aggregate [82], recycled rubber particles [99], and fired clay waste [101].

The second method is encapsulating the bacterial spores by adding them to capsules. These capsules are made of specific substances like glass, poly, and melamine. The encapsulation method was reported to be better than the direct method [56]. On the other hand, the complex procedures, professional equipment demand, and relatively high cost are the main drawbacks of the encapsulated method [99]. Also, due to issues with workability and strength, the capsules' amount is typically limited to 2% by the weight of the concrete [19]. As illustrated in (Fig. 5), the repair begins when the capsule is broken due to the crack initiation and propagation through it, releasing the healing agent, which can be chemicals or micro-organisms, filling the crack and preventing it from spreading further [32, 94].

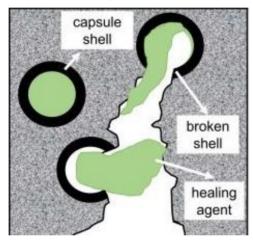


Fig. 5. Encapsulated method healing technique [19].

The inadequate design of the entire encapsulation system may allow the crack to develop around the capsule rather than penetrating it. The shell thickness also affects the performance of capsulated bacteria; using too thick shells prevents shell rupture while using thin shells allows the healing agent to be released while mixing [102]. When large-size capsules with diameters ranging from 400 to 600 μ m were used, only around 8.6 % of the capsules broke, while the remainder were moved out of the matrix, dropping voids. When smaller capsules (50–200 μ m) were used, approximately 34% were ruptured [103].

pH value

The pH of the media influences the growth and survival of bacteria. Each microbial species possesses a definite pH to help grow. The activity of bacteria decreases significantly with increasing pH value as metabolic and biomineralization activities decrease as the pH value increases [91]. Cement's high pH (between 10 and 13) makes bacterial growth in an unfavorable environment [70]. Therefore, the rise in pH is a key problem that faces bacterial concrete.

Bacteria Concentration

Cell concentration can affect the mechanical characteristics of concrete [78]. Many researchers have investigated the impact of various concentrations and observed the optimum concentration. For example, 10^5 cells/ml of mixing water was the optimum cell concentration, resulting in the best mechanical properties using Shewanella and Bacillus Subtilis micro-organisms [4, 11, 39, 43, 64, 65, 86]. Findings suggest that the hypothesis positing a correlation between the increase of bacterial concentration and the improvement of compressive strength is not entirely accurate, as evidenced by the reduction of compressive strength by 10% when Bacillus pseudofirmus was utilized at a cell concentration of 10^8 cells/ml [25, 84].

· Nutrient media

Healing ability can be influenced by various factors associated with nutrient media. The presence of this substance can enhance the formation of the healing compounds required for crack closure and promote the growth of bacterial spores [39].

The selection of such materials is critical because they must function well with concrete and not promote deterioration. In general, materials that are common to retard the setting of concrete, such as chlorides and sulfate, should be avoided. Researchers have used different nutrient media combined with organic compounds such as urea, yeast extract, sodium, protein, sugar, buffer solution, calcium lactate, and calcium acetate for metabolic pathways.

Sugars, like glucose and dextrose, have a more significant delaying influence when added after mixing the water and the cement [104]. Sugars' retarding effect is augmented when increasing sugar content. A sugar content of 0.1% by cement weight can delay hardening [105]. Using sodium as a nutrient medium was preferable [95, 106]. In addition, using sodium citrate has been shown to affect the setting and hardening of concrete [106].

Proteins, such as peptone and tryptone, were evaluated [26, 64, 84]. Strength was reduced when using 1% peptone of cement content [64]. Using large amounts of calcium lactate can postpone the setting time, decreasing the concrete's strength at an early age. Calcium lactate was the most used nutrient [10, 25, 43, 64, 89, 92] with different percentages. It was observed that 1% and 0.5% of cement content give better results than 5 and 10% in compressive strength.

Yeast extract and calcium acetate have been used as nutrient media to study their effect on concrete mechanical properties [64, 95, 107]. They found that using yeast extract and calcium acetate by 1% of cement weight reduced the compressive strength by half [64]. However, 0.5% or less of cement weight does not affect the compressive strength when using yeast extract [95, 107].

A small amount of urea (0.5% of cement weight) does not impact concrete properties or bacteria. However, 4% urea of cement weight could delay the hydration [94].

5. Self-Healing Concrete Performance

5.1. Structural Properties

Bacterial concrete's structural properties can be affected by a number of variables, such as the bacteria used and their concentration, the type of nutrient media employed, and the presence of admixtures or additives. In order to design safe, durable, and efficient concrete buildings, it is crucial to understand and optimize these properties. Table 1 shows the results of past research on mechanical properties (compressive and flexural strength) after 28 days.

Table 1. Summary of mechanical properties past research

Reference	Bacteria type	Concentration	Insertion method	Nutrient	Compressive strength	Flexural strength
[85]	B. pasteruii	-	Indirect	-	135%	-
[78]	B. subtilis	106			↑28%	
		$10^7 \& 10^4$	Direct	-	↓9%	-
[86]	B. pasteurii	10 ⁵	Direct	Silica fume (10%)	↑14% ↓6%	-
[87]	B. pasteurii	-	Direct	-	↑30%	12%
[77]	B. subtilis & B. megaterium	108	Direct	Calcium lactate (0.5%)	16%	-
[26]	B. subtilis	10 ³	Direct	Peptone	↑5% ↑42%	-
[89]	B. subtilis	109	Direct	Calcium lactate (10%)	↓21%	-
[10]	B. subtilis	108	LWA GNP	Calcium lactate (5.6%)	↑12% ↑9%	-
[80]	B. subtilis	10 ³ 10 ⁶	Direct	-	↑13% ↑21%	-
[90]	B. Sphaericus	-	Flyash		↑69%	↑110%
[,]	B. pasteurii		10%		↑37%	↑27%
[97]	Bacillus subtilis	108	Direct Iron oxide Bentonite	Calcium lactate (1%)	↑9% ↑21% Slight increase	-
[99]	Sporosarcina pasteurii	1013	Small rubber particles Large rubber particles	Calcium acetate (2%)	Slight increase	-

5.2. Microstructure

In order to investigate the morphology and microstructure of concrete, a variety of tests can be conducted, such as (scanning electron microscope (SEM), x-ray diffraction (XRD), and Energy dispersive x-ray spectroscopy (EDS)). Several microstructure tests are commonly performed on bacterial and ordinary concrete to examine the formation and distribution of calcium carbonate crystals and the overall influence of bacterial activity on the microstructure of the concrete matrix. The results obtained from these experiments contribute to assessing the efficacy and long-term resilience of self-healing concrete as a self-repairing substance. Table 2 presents a comprehensive overview of researchers' various testing methodologies to investigate the microstructural aspect. The table also provides a summary of the findings obtained from these investigations.

Table 2. Summary of microstructure tests and research findings

Tests	Reference	Objective	Main findings		
SEM	[17, 83, 108, 109]	Evaluate the changes in concrete microstructure after the self-healing process.	The control samples showed small quantities of CaCO ₃ crystals, in contrast to the bacterial concrete mixtures.		
	[6]	SEM analysis was performed on the strain solution precipitates that formed during MICP.	Samples of chemical and bacterial concrete both revealed polygonal calcium precipitation.		
	[96]	Investigate the effect of air- entraining admixtures on the bacterial concrete.	The morphology of calcium carbonate is affected when air-entraining admixture is used.		
	[82]	Examine the available evidence for any indications of biomineralizations.	Bacterial activity within concrete structures has been demonstrated through the formation of orthorhombic crystals.		
	[43, 68, 110]	Examine the presence of bacterial activity in the healed cracks.	Bacterial activity can potentially be indicated by Neddele and bouquet CaCO ₃ precipitation shapes.		
	[97, 111, 112]	Study the effect of bacteria in concrete using the indirect method (Immobilization technique)	SEM images were obtained to examine the distribution of Bacillus subtilis spores in nutrition broth within the chosen immobilizer.		
	[67, 113- 115]	Study the healing products' morphology, crystal phases, and chemical composition.	The results of SEM imaging on samples of varying ages do not show significant differences in the size, shape, and distribution of the crystals.		
XRD	[6, 82, 83, 97]	Study the precipitated material resulting from bacterial activity is CaCO ₃ .	CaCO ₃ was found in the healed area with C-S-H, confirming both self-healing techniques.		
	[111]	Examine the hypothesis of the continued hydration and bacterial activity.	The finding supports the hypothesis that sodium silicate underwent a reaction with the pre-existing Ca(OH) ₂ , resulting in the formation of additional C-S-H gel.		
EDS	[82, 84, 114]	Examine the presence of the CaCO ₃ compounds in the healed cracks.	Calcium, oxygen, and carbon were found in the healed areas, confirming CaCO_3 preceptations.		

5.3. Durability

The concept of "concrete durability" relates to the ability of concrete structures to withstand various environmental and operational circumstances without significant degradation or loss of functionality over an extended duration. This section will examine various factors that contribute to the durability of concrete. Several researchers have indicated that incorporating self-healing concrete can effectively improve durability. Table 3 provides a comprehensive overview of various research investigations related to the durability of concrete.

Table 3. Summary of research investigations on concrete durability studies.

Durability aspect	Bacteria type	Reference	Objectives	Main findings
	Micrococcus - B. subtilis	[116]	Investigate the effectiveness of the treatment using calcinogenic bacteria.	Capillary water absorption tests showed that calcinogenic bacteria reduced porosity by 60%.
	B. sphaericus	[117]	Investigate the impact of surface porosity on treatment efficacy.	The degree of porosity affects the water absorption.
	B. megaterim - B. cereus - Lysinibacillus	[118]	Examine how FA and different bacteria types affect biomineralization.	The porosity decreased by 24-31% due to void filling and pore-clogging caused by CaCO ₃ precipitation.
D	B. sphaericus	[115]	Examine how bacteria affect mortar porosity.	The porosity was reduced by 50% using bacteria.
Porosity	B. megaterim	[119]	Reviewing the future direction of the bacterial concrete.	Bacterial concrete reduced the porosity by 31%.
	B. cereus	[120]	Investigate the effect of bacteria immobilized in metakaolin.	The process of MICP resulted in a decrease in the porosity of mortars at all ages.
	B. subtilis	[81]	Evaluate how bacteria improve shotcrete.	The compressive strength increases as porosity decreases.
	B. subtilis	[121]	Investigate the performance of bacteria on concrete.	Using B. subtilis reduced the porosity by 70%.
	B. sphaericus	[117]	Investigate the impact of surface porosity on treatment efficacy.	Water absorption was reduced by 65 to 90% due to CaCO ₃ deposition.
	Sporoscarcina pasteurii	[86]	Investigate the impact of bacteria on the concrete matrix.	This bacteria strain reduced water absorption four times.
	-	[111]	Investigate the effect of sodium silicate on concrete performance.	The solution reduced the capillary water absorption by approximately 50%.
Water absorption	Ureolytic bacterium	[122]	Examine the effect of bacteria with baghouse filter dust.	Adding bacteria strain reduced the water absorption.
	Sporosarcina pasteurii [123]		Investigate the effect of bacteria on water absorption.	The maximum reduction in water absorption over control samples was 80–85%.
	B. megaterim	[124]	Investigate the effect of bacteria on concrete performance.	Water absorption was reduced by 5.25 and 7.35% after 7 and 28 days, respectively.

	[125]	Investigate bacteria	It was found that the water
-		effect on durability absorption was dec	
		studies.	in all bacterial mixtures.
	[86]	Investigate the impact	Using this strain of
Bacillus		of Sporosarcina	bacteria reduced chloride
Paseturii		pasteurii bacteria on	permeability from Low to
		the concrete matrix.	Very Low.
Bacillus sp. CT-	[126]	Examine how Bacillus	Using bacteria reduced the
		sp. bacteria affect	chloride ion permeability
		durability.	from (3177C) to (975.3C).
	[119]	Explore bacterial	The chloride permeability
B. Sphaericus		concrete uses and	class was decreased from
		methods.	Moderate to Low.
D		Explore how bacteria	The chloride permeability
	[92]	can improve concrete	class was decreased from
Pseudonrmus		durability.	Moderate to Low.
	[121]	Study the effect of	The chloride permeability
B. Subtilis		Bacillus subtilis strain	class was decreased from
		on the concrete.	Moderate to Very Low.
	Paseturii Bacillus sp. CT- 5 B. Sphaericus B. Pseudofirmus	Bacillus Paseturii Bacillus sp. CT- 5 Bacillus sp. [126] Bacillus sp. CT- [126]	Bacillus Paseturii Bacillus Paseturii Bacillus Paseturii Bacillus Paseturii Bacillus Paseturii Bacillus Paseturii Bacillus sp. CT- 5 Bacillus sp. CT- 5 Bacillus sp. CT- 5 Bacillus sp. Examine how Bacillus sp. bacteria affect durability. Explore bacterial concrete uses and methods. Explore how bacteria can improve concrete durability. Study the effect of Bacillus subtilis strain

6. Summary and Conclusions

This study has identified two approaches to self-healing concrete (autogenous and autonomous) and the influence of utilizing bacterial concrete. Autonomous self-healing was found to be more effective than autogenous self-healing. Bacterial concrete exhibits notable outcomes in terms of permeability and strength due to the presence of bacteria. Based on the current research, bacterial concrete is becoming known as an environmentally friendly alternative. At some point, it will improve the longevity of the materials used in construction.

- After 28 days, it has been observed that approximately 30% of the cement particles present in conventional concrete remain unhydrated. Even though, the hydrated cement particles can have an inner core that remains incompletely hydrated, requiring further hydration over time. This phenomenon plays an essential part in improving the autogenous self-healing mechanism.
- Fly ash and silica fume are two examples of pozzolanas that have a beneficial effect on autogenous healing because of their delayed reaction with calcium hydroxide. However, using excessive amounts of FA can decrease compressive strength.
- Calcium hydroxide and calcium carbonate formation exhibit superior autogenous self-healing properties compared to calcium-silicate-hydrate (C-S-H).
- The use of bacteria in self-healing concrete can prevent steel corrosion, reduce permeability, and enhance mechanical properties, making it more durable and substantial.
- Autonomous self-healing concrete has resistance to freeze-thaw and high carbonation, which can help decrease the porosity and permeability.
- There is no relation between compressive strength and bacterial concentration.
- Eurocode 2 is the first code to allow the use of capsules in applying bacterial concrete.
- The characteristics of microbial precipitation can be affected by several aspects, including calcium concentration, pH value, dissolved inorganic carbon concentration, and nucleation sites.

- The efficacy of the indirect approach was found to enhance healing ability to a greater extent than the direct method. This is attributed to the prolonged viability of bacteria achieved through the indirect approach.
- The particle size of immobilizers influences the rate of healing. Since, particles with a larger size exhibit superior performance due to their ability to seal cracks effectively. Moreover, it has been observed that larger particles possess a greater ability to immobilize bacteria compared to smaller particles.
- It is preferable to use small capsules rather than larger ones since large capsules can rupture while mixing or hardening.
- Water, gas, and chloride permeability can be considered self-healing indicators.
- Nutrients are an essential parameter that aids bacteria to survive and help in self-healing concrete. In addition, the negative impacts of nutrients can be controlled.
- The most familiar cause of self-healing in both techniques is the precipitation of calcium carbonate crystals (CaCO₃) in the crack.

7. Future Recommendations

Several suggestions are made for future research. These suggestions are not meant to be exhaustive, but they do cover much ground and could include (but are not limited to) the following:

- Conducting further studies to explain better the adverse effect of using different immobilizers for bacterial concrete and its effect on mechanical properties.
- Studying the effect of different strains and higher concentrations of bacteria, alternative feeding agents, and various immobilization techniques on concrete.
- Future research should investigate the feasibility of implementing self-healing mechanisms in structures submerged in marine environments.
- Conducting experiments on larger models or prototypes is crucial in facilitating this novel technique's widespread acceptance and implementation.
- In order to assess the economic viability of incorporating a bio additive in concrete, particularly in the context of construction materials, it is imperative to conduct feasibility studies.
- The implementation of standardized protocols is recommended for evaluating the compatibility of bacteria with construction materials, specifically in determining the optimal bacterial concentration to be incorporated into concrete and the corresponding water-to-cement ratio for different applications.

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