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Review of autonomous self-healing cementitious material

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Abstract. Concrete is a well-known versatile material, and its application is very common in most structures. Concrete performance is high in compression but low in tensile strength, this leads to the appearance of microcracks when the structure bears the designed loading. Such microcracks when ignored, leaves the structure vulnerable to attacks such as seepage of water, chlorides, and other materials that lead to a reduction in performance, and extreme cases failure of the structure. Since cracking is inevitable in concrete, new materials with self-healing properties are introduced into the mixture to take advantage of the external materials while making the concrete stronger. This type of concrete is widely researched from 1970 until the present day and is still in 'proof of concept stages, and very few to no applications of autonomous self-healing concrete in real-world structures. This paper is an attempt to further classify the existing methodologies and find the gaps between researchers. The autonomous healing of concrete in present-day research varies in results; this means that the self-healing methodology requires standardization. Furthermore, self-healing in concrete does not mean maintenance is not required, it implies an easier maintenance method is possible due to the benefits gained through a possibly higher early cost in construction.

1. Introduction

Following economy 4.0, the concrete industry also develops "Smart" concrete with innovations such as self-compacting concrete, translucent concrete, aerated concrete, 3D printed concrete, lightweight concrete, and even self-healing concrete. Concrete is a commonly used material when compressive strength is required, although such an impressive material also has flaws. Designed loads applied to concrete may not be a problem, but new consequences arise when certain circumstances force the load to be higher than the designed compressive strength of the material. Cracks may start to appear due to tension, and it can cause some reduction in its durability, strength, and permeability. The concern for cracks is due to the penetration of harmful agents such as CO₂, SO₄²⁻, and CL⁻ that causes corrosion in steel rebar in the presence of water and oxygen in concrete [1,2]. Such problems lead to innovations in improving the overall performance of concrete, through aspects of sustainability, such as using renewable and cost-efficient materials, optimization for production processes, and even new construction methods.

Concrete is known to have low tensile strength, and in most cases, concrete is prone to crack [3]. The degree of severity in cracking depends on human error, in some cases, it is due to poor detailing, design errors, and even construction errors. The workmanship skill of construction workers plays a strong role, and previous researchers found that in most cases, the uncontrolled addition of water when pouring insitu concrete is an actual problem in Indonesia and the United Kingdom [4-6]. Such unwanted errors cause problems in concrete, and it means that the poured concrete requires further rehabilitation, which



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may cause additional expenses [7]. Several researchers have made solutions that made it probable for automation of concrete rehabilitation. Previous researchers also stated that maintenance cost is highest at 39.88% in building maintenance issues [7]. This further supports the idea, and potential benefits of self-healing concrete when applied in construction.

Concrete on its own is a quite durable material, but its performance can decrease throughout time due to multiple factors, leading to a shortened service life. It is possible to develop methods that can prolong the service life of concrete using self-healing techniques, as seen in Figure 1.



Figure 1. Performance vs. service life of concrete (adapted from [8,9]).

2. Mechanism of self-healing concrete

Combating cracks by using concrete alone is not enough; and to achieve significant optimization, the use of other materials is essential. Each alternative has different capabilities in remediating concrete cracks. There are two main categories of technological advancement in self-healing concrete, as follows:

- Natural/autogenous healing: Previous researchers have proven that the penetration of air and moisture in the concrete allows the non-hydrated cement particles to react. Such a reaction produces the self-healing effect and fills micro-cracks that occur within the concrete material [10–14].
- Engineered/autonomic healing:
 - Modified Autogenous healing: New advancements using other cementitious materials such as the use of microorganisms, fly ash, fibers, polyvinyl alcohol, fine silica, and even utilizing the Calcium–Sulfate–Aluminate reaction to fill cracks [2,14,15].
 - Encapsulation of healing agents: This method is popular and effective in combating concrete cracks compared to the other methods. Using microcapsules to immobilize bacteria allows it to be dormant within the concrete until needed. When the concrete cracks and breaks its capsules, the healing agents begin to fill the cracks [16–18].
 - Vascular methods: This system adopts the human cardiovascular system as blood transports food, water, and minerals throughout the body. When applied to concrete, it can deliver healing agents from one point to another from an external source within the network [19–21].

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The different techniques in the application of healing agents are compiled and are as seen in Table 1, Table 2, and Table 3 respectively for modified autonomous healing, encapsulation, and vascular methods.

The three methods mentioned in the tables above have explored multiple variations and contribute new results to the self-healing materials category. Several concerns arise as the self-healing capabilities of one researcher vary in size. This is due to the pre-cracking of concrete that produces a variety of crack sizes as well. Nugroho et al. 2015 discovered a stiffness recovery of 34.85% of the original flexural strength, although, permeability tests reveal that precipitation of calcite erodes when crack sizes above 0.22mm even after it is filled [22]. Renewed methods using popular powder incorporated directly into the concrete mix with some additives of Mn^{2+} , Fe^{2+} , and Ca^{2+} reveal a higher healing capability up to 0.33 mm average, however, no permeability tests were reported [26].

Upon inspection, cracks that are healed by not relying on hydration are minuscule in their results. Cracks from 100 micrometers up to 0.33 millimeters on average are the threshold for the healing agents to cover. The drawback of the healing products is its unreliability in strength since it is brittle and can only cover static cracks. Most of the healing methods require the presence of water to initialize the healing process, and in extreme cases require a full submersion. Furthermore, bacteria-based healing requires not only the bacteria itself but also enzymes that act as catalysts to produce the healing product. The above reasons limit the healing process of concrete, not to mention that the healing agents are still uncommon in practice and can further add costs to the concrete [38].

Source(s)	Specimens	Techniques	Remediation Result
[22]	<i>Bacillus subtilis</i> induced specimens varying in cell concentrations of 10 ⁴ , 10 ⁵ , 10 ⁶ cells/ml.	Direct mixture into the cement matrix Compressive Strength Flexural Strength Permeability test X-Ray Diffusion tests	34.85% of its original stiffness was regained with 10 ⁶ cell/ml solution. Permeability tests revealed complete recovery of 0.22 mm crack healing, and 10 ⁵ cell/ml is most effective.
[23]	<i>Sporosarcina pasteurii</i> isolated and grown in urea and phosphate bugger at 10 ⁸ and 10 ⁹ cells /ml concentrations	Direct mixture into the cement matrix Compressive Strength Scanning Electronic Micrographic	Results show a 24.9% increase in compressive strength and 37% in tensile strength
[24]	<i>Bacillus subtilis and Bacillus megaterium</i> at 10 ⁸ cells /ml	Direct mixture into the cement matrix Compressive Strength Split Tensile Strength Scanning Electronic Micrographic	Both types of <i>Bacillus</i> bacteria revealed a 14.3% and 22.5% increase in compressive strength, and an increase of 25.3% and 15.8% in tensile strength for <i>Bacillus subtilis</i> and <i>Bacillus</i> <i>megaterium</i> respectively
[25]	<i>Bacillus subtilis</i> included in specimens varying in concentrations of 10 ³ , 10 ⁶ , 10 ⁹ cells/ml.	Direct mixture into the cement matrix Permeability test Ultrasonic Pulse Velocity	Increase of compressive strength and reduction of water absorption after loading reported most

Table 1. Techniques and remediation results of modified autonomous healing in concrete.

Source(s)	Specimens	Techniques	Remediation Result
		Compressive Strength Scanning Electronic Micrographic	effective at 10 ⁶ cell/ml dosage
[26]	Use of malt powder, rice bran, (NH ₄) ₂ SO ₄ , and corn syrup enhancing growth of bacteria <i>Lyisinibacillus</i> <i>boonitolerans</i>	Direct mixture into the cement matrix Visible crack width observation	Complete recovery up to 0.33mm cracks healing effect in 7 days. Mn, Fe, and Ca contributes to the sporulation of bacteria, ensuring a higher survival rate.
[27]	5 samples of microorganisms from the <i>Bacillus</i> family were stripped down to their RNA and tested for microbial growth using 2 different nutrients of Yeast Extract + Urea and glycerol added to the latter.	Direct mixture into the cement matrix Compressive Strength	Bacillus cereus or Bacillus thuringiensis as the microorganism showed an increase in 14.2% compressive strength. The addition of glycerol acted as a catalyst in cell growth at 4.23×10^7 cells.

Table 2. Techniques and remediation results of encapsulation of healing agents.

Source	Specimens	Techniques	Remediation Result
[28]	Diatomaceous earth as vessels for bacteria <i>Bacillus</i> <i>sphaericus</i> embedded in concrete specimens	Diatomaceous earth as a medium for bacteria. Permeability testing	0.15 – 0.17 mm cracks partially filled
[29]	Analytical model for determining microcapsule dosage.	Mathematical equations for 2D and 3D models	The self-healing matrix is purely dependent on the occurrences and probabilistic simulations of crack formations.
[30]	<i>Bacillus sphaericus</i> in a medium of liquid minimal basal salts kept at 10 ⁹ cells/ml concentration	Microcapsules embedded in mortar mix Compressive Strength Test Permeability test Pore size distribution test	Enhancement in self-healing is visible in concrete specimens containing bacteria microcapsules. Without wet-dry cycles, healing is not apparent in the specimens.
[31]	Concrete beams of 150 mm x 250 mm x 3000 mm with glass capsules embedded inside containing superabsorbent polymers (SAP) and Polyurethane (PU)	Capsules are wired and placed using wires. LVDT measurements in tensile zones x-ray microtomography	Cracks were repaired partially, but healing performed better with the healing agents when compared to the reference beam. Permeability was not able to be tested

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Source	Specimens	Techniques	Remediation Result
[32]	<i>Bacillus subtilis</i> is used at concentration of 2.8x 10 ⁸ cells /ml	Direct mixture into cement matrix using lightweight aggregates (LWA) Compressive Strength X-Ray Diffusion tests	Healing is evident in concrete early ages of 3-7 days and decreases in later ages. Lightweight aggregate deemed inefficient as a medium. Compressive strength increased in comparison to reference concrete specimen.
[33]	Healing over damage/healing cycles of specimens containing <i>bacillus sphaericus</i> of 10 ¹⁰ CFU/ml combined with Superabsorbent Polymers (SAP)	Compression Strength Permeability test Visible crack width measurement Scanning Electronic Micrographic	The addition of SAP and bacteria fills cracks in concrete even after repetitive loading tested in three damage/healing cycles and performs better than the reference concrete.

Table 3. Techniques and remediation result of vascular system distribution.

Source(s)	Specimens	Techniques	Remediation Result
[34]	Concrete beams equipped with hollow fibers containing adhesive methyl methacrylate	Use of heat in releasing healing agent within the tubes into concrete	Flexural capabilities increase for concrete equipped with adhesives.
[35]	Concrete test beams and columns were given loading while adhesive- filled fibers were distributed along the axes.	Three-point bending test and assessed after damage/healing cycles	Ductility improved for samples containing adhesive. Bend test proved higher than the reference concrete.
[36]	Capillary tubes embedded in the tension zone of a concrete beam containing cyanoacrylate adhesive.	Concrete beams gave a 5mm notch for testing	Self-healing is evident but limited due to the unevenly broken tubes within the tubes.
[37]	Hollow fiber tubes embedded in the tension zones beneath steel reinforcement acting as a vascular network for the distribution of healing agent	Three-point bending test	Improvement in strength and stiffness of concrete, like the tubes, enable healing agents to be supplied continuously.

3. Real-world applications of bacterial concrete

The above-mentioned methods show potential in self-healing concrete. Although modified autonomous healing and encapsulation of healing agents are more popular compared to the other two techniques

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among researchers, it is also necessary to estimate its feasibility from both industrial and economic points of view. The maintenance cost of conventional concrete contributes nearly 40% of problems enlisted by experts [7,20]. New maintenance techniques have been proposed using nondestructive tests, including ultrasonic and acoustic emission techniques, although it is stated as both times consuming and requires high labor, and can potentially add to the maintenance costs [39]. Broadening the topic on cost, the life-cycle cost of self-healing in concrete is difficult to estimate, and very few applications exist in real-world structures [37]. It is estimated that the additional self-healing properties using polyurethane and *bacillus subtilis* may increase concrete costs from 7-28% [40]. This means that costs in construction may increase, and the maintenance costs decrease, however, estimates of its value is still unclear. The popular encapsulation method cost estimate at around \$15 /kg consisting of urea-formaldehyde as shell and epoxy resin as its core [41]. Large-scale laboratory testing is scarce, and field applications of self-healing effects require further proofing. Some large-scale tests and field observations are listed in Table 4.

Source(s)	Specimens	Techniques	Remediation Result
[42]	Improvement of freeze- thaw resistance by liquid- based systems on 8-year- old garage using <i>Bacillus</i> genus bacteria	Core drill tests Permeability tests	Mass loss of concrete material is visible, nearing half of its original weight. And permeability tests reveal better performance using bacterial liquid treatment.
[43]	 The researcher gave an overview of self-healing applications using <i>Bacillus</i> genus bacteria in; Irrigation canals in Ecuador, Patch repairs using self-healing mortar in cracked walls. Improvement of freeze-thaw resistance by liquid-based systems on the 8-year-old garage. 	 Encapsulation of bacteria in lightweight aggregates was used in the creation of new irrigation canals. Patching was done with both normal cement mortar, and bacterial mortar, by cutting open the cracked wall, and filling it with the new mortar. Bacillus genus liquid with solutions; sodium silicate, sodium gluconate, bacteria, calcium nitrate 	 5 months of use and cracks are not visible, and no signs of deterioration were detected in canals 6 months after repair, normal cement mortar is discolored, although no delamination is visible for both mortar types. Leakage is minimum with the use of bacteria when compared to untreated cracks.
[44]	 Upscale testing using Bacillus genus bacteria in: A concrete beam of 150x250x3000 mm3 was made with the addition of encapsulated bacteria in diatomaceous earth (DE), and hydrogels. Irrigation canals made using lightweight aggregates (LWA) coated with a bacterial solution 	 Four-point loading was applied and water supply was given in at intervals. Permeability tests were also conducted. Compressive strength test, three-point bending test, visual crack sealing observation Compressive strength, flexural strength, and permeability test 	 Bacterial concrete proves to have better water retention properties, although water permeability healing was not significant. The use of hydrogels resulted in drastic strength loss (50%) The test was sufficient, and resulted in the application of the canal, cracking n=or deterioration is not present one year after casting

Table 4. Large-scale tests and field observations of self-healing.

Source(s)	Specimens	Techniques	Remediation Result
	- 16 m2 concrete wall repair using LWA coated with a bacterial solution		- The laboratory specimens significantly performed better, but field application results were not reported.
[45]	Creation of a small-scale slab cast with bacterial self- healing concrete	Compressive strength for cube specimens. Permeability test under wet/ dry cycles within a large pit made especially for slab inspection	Real scale roof slab was yet to be applied, although the test reveals no condensation on the bottom side of the lab in the early stages. Upon inspection one 1 year after casting, condensation occurs, and it is concluded that direct contact with water is required for significant healing

4. Unexplored territories and further improvements

The precipitation process of bacteria has many applications, but many methods require assessment in its feasibility. Six criteria's are provided to ensure the robustness of a self-healing method are as follows [38,46]:

- [1] Shelf life: Concrete itself has a relatively long service life, meaning that any self-healing materials should also possess such criteria.
- [2] Pervasiveness: The damages that may occur anywhere within a concrete structure and selfhealing should not be limited to discrete parts.
- [3] Quality: Complete recovery of damages is important so that mechanical properties including stiffness, strength, and ductility remediation is essential. This means the filling of the crack is important, but the bondage is inspected.
- [4] Reliability: Self-healing consistency in delivering its remediation capabilities still vary and applications are scarce.
- [5] Versatility: Environment plays an important role in the durability of a structure; self-healing that is capable to recover material properties in various environments is required.
- [6] Repeatability: Indeed, long service life is common for a structure; damage is also a repeatable occurrence. This implies that the remediation process must remain active in multiple damage/healing cycles.

The number of publications regarding self-healing is rising exponentially since Malinskii discovered the development of crack healing using polymeric materials in 1970. In recent years; the remediation of toxic metal waste has been a topic of interest regarding bacteria [47]. The pursuit of knowledge for self-healing technology continues to rise but applications are scarce since it is still in the 'proof of concept' stages [30,37]. Previous researchers stated that treatments are limited due to the lack of commercial production and practical engineering experience in microbial self-healing concrete [48].

It is proven on a laboratory scale test, that precipitation of CaCO₃ maybe dissipate due to water flow in the cracks [22]. The organic characteristic of urea is thought to be the problem, and the use of other cementitious materials may be explored to lessen the weakening effect. Otherwise, exploration of other uses of bacterial mortar such as plaster may be a more viable approach [49].

There are some unexplored areas in self-healing concrete technology. It is known that some bacteria are temperature resistant, enabling them to survive the high temperatures within concrete due to hydration. Higher temperatures occur in mass concrete, and the survivability of bacteria in such temperatures requires observation. Another area of research concerns the damage/healing cycles of concrete, as laboratory conditions could be adapted to real-world conditions. Cyclic loading is another topic of interest since concrete endures not only environmental attacks but also such loads. Studies on

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cyclic loading on self-healing materials are relatively new as the applications of self-healing are still in their 'proof of concept stages. Early results reveal a positive impact of self-healing materials both in bearing capacity and the number of cycles endured [50,51]. New monitoring techniques could be developed for self-healing, past studies have utilized the use of acoustic emission, elastic wave tomography, electrochemical impedance spectroscopy, and ultrasound pulse velocity measurements [39,52–54].

To increase maximum crack-width healing capacity, while also increasing concrete properties, selfhealing materials need to be adjusted according to the six above-mentioned criteria. Such as the survivability of healing agents under acidic conditions, or in near-shore environments using materials that are low in cost. Fermentation and drying processes that are low in cost can be considered in obtaining bacteria for future production strategies in the self-healing industry [38].

5. Concluding remark

The review examined by previous researchers is relevant and plays a strong role in future perspectives of different self-healing concrete applications. Autonomous healing in concrete designed with mixtures of bacteria, certain minerals, and polymers offers benefits such as better performance compared to conventional concrete. However, the self-healing methodology does not have standards yet; further analysis on present methodology regarding shelf life, pervasiveness, quality, reliability, versatility, and repeatability is important to ensure its robustness. Future applications also require thorough maintenance procedures, since most present methodologies ignore real environmental conditions.

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