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Review

Factors influencing self-healing mechanisms of cementitious materials: A review

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A B S T R A C T

The increasing awareness of climate change and global warming has pushed industries to be more conscious of their environmental impact, especially in the construction industry with the main contributor being concrete. Concrete is a material that is in very high demand in the construction industry for structural applications. However, it's a material with a major concern with the challenges of microcracking. New technology has seen the development of self-healing material, using novel techniques to bring cementitious materials back to its original state. This paper reviews and evaluates the novel techniques adopted by the researchers in the field to achieve a self-healing material, with the main focus being on the factors influencing the mechanisms of autogenous healing and bacteria-based healing. Various parameters including bacteria type, pH, temperature, nutrient, urea, and Ca^{2+} concentration, bacteria concentration and application, pre-cracking, healing condition, cement type, and crack width are all important for healing efficiency, although the use of water to facilitate both autogenous and ureolytic bacteria healing mechanism is paramount for the triggering of healing processes. This study thoroughly presents various factors and their correlation to the healing mechanisms of autogenous healing and ureolytic bacteria healing. Further studies are identified to better understand the exact mechanism taking place and which healing process contributed to how much of the healing, and this review could serve as an informative platform for these pursues.

1. Introduction

Concrete is a material that is in very high demand in the construction industry for structural applications [1]. It is the second most utilized substance on the planet after water and the most utilized man-made material [2–4]. Such popularity, especially as a structural material, is due to its favourable properties, including high compressive strength, non-combustible nature, mouldability that allows for flexible design, availability and low costs of raw materials [5]. One of the main disadvantages of concrete is its very low tensile strength, which is practically exceeded at low levels of load. This results in the cracking of concrete surfaces that in turn leads to various aesthetical and structural problems. Reinforcing steel has rather high tensile strength and a symmetrical material constitutive law under tension and compression.

De Belie and Jonkers found that microcracks formation of up to 0.30 mm in concrete is inescapable [6]. These microcracks can form before or after the hardening of the concrete. The former usually occurs due to physical, chemical, thermal, or structural causes, whereas the latter occurs due to plastic cracks (shrinkage, settlement, autogenous shrinkage) or construction movements (e.g. tensile stresses, freeze thawing) [7]. Although microcracks of up to 0.30 mm aren't generally affecting the overall structural integrity, these small openings can be detrimental, as the cracks will allow for harmful substances to possibly deteriorate the concrete and cause oxidation of the steel reinforcement which is dependable on the extent of corrosion and deterioration and can potentially cause catastrophic failures if left untreated. Ultimately these microcracks can lead to a decrease in the strength and durability of concrete structures due to permeability issues if not addressed.

Several alternative solutions are nowadays available on the market to mitigate some of the issues related to concrete surface microcracking. These solutions include: (i) grout injection directly into the crack using a pump [8], (ii) applying epoxy resin grouting [8], (iii) over-sizing members [9], and finally, (iv) applying mortar over the cracked surface. The common denominator of these solutions is to block any new

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harmful substances from entering the concrete microstructure and to avoid further human intervention. However, the listed solutions don't prevent the oxidation of the steel reinforcement already in place from stopping. Hence, the need for a solution that will target the root cause of the problem and actively prevent these harmful substances from advancing within the structure where they can cause significant complications is required.

An alternative and more effective solution for the repairing of concrete structures is represented by dispersing polymers[10], fibres[11], bacteria [12–16], chemical healing agents [17–19], and mineral admixture [20], inside the concrete batch to turn concrete into a self-healing material or as a surface treatment of existing structures [21–23]. Self-healing is the ability to detect and repair damages in the form of cracks without the need for human intervention [24]. Self-healing can be known to be a biomimetic process, where the concrete healing mechanism mimics that of human tissue being repaired after a wound. Using this, analogy, self-healing concrete resembles a human body that is capable of detecting damage and healing itself to some degree based on several factors including the depth and width of the wound. This review paper will discuss the latest research findings on self-healing concrete and critically analyse the factors potentially influencing the healing properties. As the construction industry is moving towards a more innovative and sustainable route, self-healing concrete can aid in reducing industry-related environmental impact and the financial burdens associated with the production and dismantling of concrete and concrete systems. As ordinary Portland cement (OPC) amounts to 7% of global CO_2 emission produced by humans (Jonkers et al., 2010), self-healing concrete provides a solution to reduce this by preventing the need to produce more concrete as the service life of structures is improved. The objective of this state-of-the-art literature review of factors influencing the autogenous healing mechanism and bacteria-based healing mechanism is to show factors which can promote or hinder the healing process, and provide a better understanding of how to go forward with improving the healing process with advanced developments.

2. Autogenous healing mechanism

According to De Rooij *et al.* [24], autogenous healing can be defined as a healing process that occurs and is triggered by components originally found in the material without further external additives or human interference. Since the material isn't designed for self-healing but it happens naturally, autogenous healing is often referred to as a natural healing process. Hearn, 1998, [25] defined it as a natural process in which cracks are amended in the presence of moisture and absence of stress. Autogenous healing can take place in favourable conditions such as the presence of water and temperatures within range to promote reactions to take place [24,25]. Several research articles suggested four mechanisms [24,26,27] grouped into three different causes that promote the occurrence of autogenous healing in concrete structures, as depicted in Fig. 2. The first mechanism has a physical cause linked to the swelling of the cement paste located near the faces of the cracked surface [24,27]. This occurs as a result of the propagation of a crack and the consequent penetration of water into the crack, which is absorbed by the hardened cement paste and causes it to swell and expand. The resulting gradual expansion seals the crack when the two surfaces of the expanded crack form a bridge bond. However, De Rooij *et al.* [24] considered this mechanism ineffective, reporting that the effect of swelling isn't substantial and only reduces the fluid flow by <10%. This suggests that although this mechanism occurs in cracked concrete, it cannot equate to very high healing, which would imply the ability to heal deep cracks is insufficient. The second mechanism is linked to a chemical cause related to the continued hydration of cement particles within the cementitious material matrix as a result of cracks allowing water to enter and react with the anhydrous cement as illustrated in Fig. 1. Water triggers the healing mechanism where dry cement takes up the space forming calcium silicate hydrate gel (CSH) [24,27,28] as well as other hydration products such as calcium hydroxide, calcium sulphoaluminate, and aluminates [24]. However, continuous hydration is limited being able to only heal cracks up to 0.02 mm wide and therefore cannot be considered a significant mechanism for repairing larger cracks [24].

While in the chemical healing causes, a second mechanism is represented by the formation of calcium carbonate ($CaCO_3$) illustrated in Fig. 1. This mechanism was discussed in several studies to be the primary mechanism found in autogenous healing [24,27,28]. Calcium ions (Ca^{2+}) available in the cementitious materials matrix are responsible, together with water penetrating the pores along with CO_2 entering as a result of the crack, to react and form carbonate ions (CO_3^{2-}). The available Ca^{2+} reacts with the CO_3^{2-} in the water to form calcium carbonate precipitate ($CaCO_3$). The reactions can be summarised as follows [28]:

$$H_2O + CO_2 \leftrightarrow H_2CO_3 \leftrightarrow H^+ + HCO_3^- \leftrightarrow 2H^+ + CO_3^{2-} \tag{1}$$

$$Ca^{2+} + CO_3^{2-} \leftrightarrow CaCO_3(pH_{water} > 8) \tag{2}$$

$$Ca^{2+} + HCO_3^- \leftrightarrow CaCO_3 + H^+(pH_{water} > 8) \tag{3}$$

Eqs. (1)–(3) show the formation of $CaCO_3$ on the crack surfaces, which in turn helps gradually seal the crack. The last mechanism is due to physical causes linked to loose debris as a result of the cementitious material cracking or fine impurities found in water [24,26–30]. However, to benefit from this mechanism, the water pressure should be high enough to move the free particles and place them appropriately and seal the crack. Therefore, this mechanism is less likely to take place compared to the other mechanisms in real-world applications. In fact, several authors, such as Ramm and Biscoping [29], De Rooij *et al* [24], argued that the aforementioned mechanism is considered irrelevant and excluded it from the list of possible autogenous self-healing mechanisms.

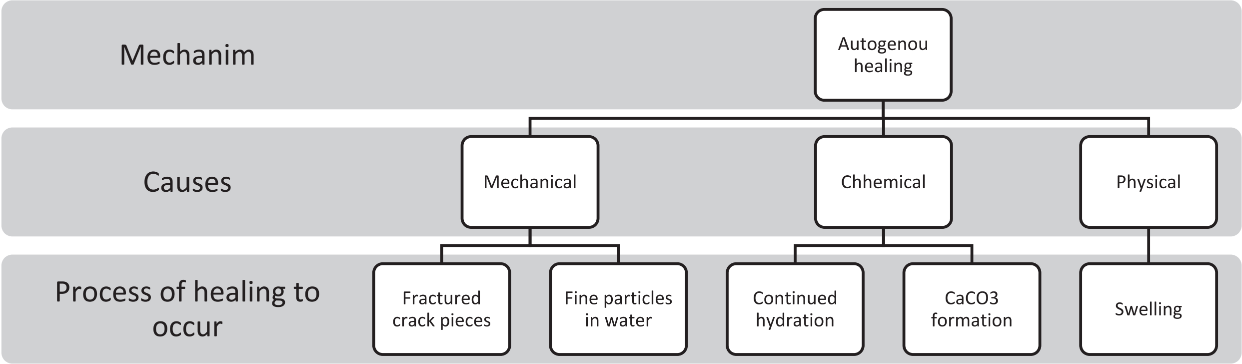


Fig. 1. Diagram showing the various processes of healing by anautogenous healing mechanics and their causes.

3. Factors influencing autogenous healing

Several influencing factors have been identified by researchers which affect healing mechanisms in concrete. These factors may have positive or negative influences, not only on the healing capabilities in cementitious materials but also on their global mechanical properties. The following sections will identify and critically discuss the influencing factors hindering or promoting autogenous healing mechanisms.

3.1. Effect of pre-cracking age

Pre-cracking age was studied by several researchers in the past [31–36], and it was found that the pre-cracking age has a significant influence on not only the type of autogenous healing mechanism taking place but the healing capabilities as well. Yang *et al.* [31] studied pre-cracked specimens which have been cured for six months by exposing them to 10 wet-dry cycles to heal, with specimens being in water for 24 h and then exposed to room conditions as the dry condition. The experimental investigation was carried out using EDX analysis on a sample of the healed product retrieved from the crack. A strong peak for Ca, characterised as calcium carbonate crystals ($CaCO_3$), was observed suggesting that in the mature specimen the formation of $CaCO_3$ promoted a self-healing mechanism to take place. Similar findings were observed by [32,33], where they conducted an extended experimental investigation to compare whether a similar healing mechanism occurred in mature (≥28 days) and young (≤3 days) specimens. The author observed that in mature specimens the healed product was mostly $CaCO_3$ after 28 days, 3 months, and 8 years of testing, however in younger specimens ongoing hydration was identified as the primary cause of healing [32,33]. Yildirim *et al.* [34] investigated the mature specimens of one-year old and were able to conclude that the main healing product was confirmed to be polymorphs of $CaCO_{3,}$ with the presence of minor hydration products which was further confirmed by XRD, SEM, and thermal gravimetric analysis (TGA).

Several researchers also widened the parameters considered during the lab investigations by including control specimens and the age at which samples were pre-cracked [14,35,36]. According to those lab campaigns, 3-day crack specimens showed healing due to the continued hydration process [14]. The findings were attributable to as time progresses there will be fewer unreacted cement particles available for ongoing hydration reaction, hence for mature specimens of 28 days cured or older, healing occurs through the formation of $CaCO_3$ precipitate. Both De Belie *et al.* and Yang *et al.* observed that $CaCO_3$ was weaker than CSH [6,31]. Thus, when mature specimens were re-loaded the crack lines propagated through the healed product [31]. As a consequence, the weaker $CaCO_3$ is formed rather than the CSH gel which is attributed to giving cementitious materials its mechanical properties. This was more evident in the experimental campaign carried out by Khushnood et al., 2020, where the 3 days old control specimen was able to regain an estimated 62% of its original strength, whereas 28-day old specimens were only able to regain 57% of its strength [35]. Similarly, in an investigation by Shaheen et al., [36], an opposite correlation was observed, as the pre-cracking age of the specimen increases, the strength regained of the specimen decreases. Most research in this field focused on proving that healing products were able to form in the cracks, however according to the authors' knowledge, fewer researchers tried to analyse the properties of the healing product as done by Yang et al., [31]. During testing of the mechanical properties, the authors identified $CaCO_3$ to be an area of weakness once the specimens have healed, however, due to limited research in this area more research is required to have a better understanding of the mechanical properties of the healed products in comparison to the other regions of a concrete (original product). This area of research would benefit from nanostructure testing of the healed products and the cement matrix to provide evidence of the possible weakness the $CaCO_3$ causes.

3.2. Effect of healing conditions

Healing conditions are known to play a vital role in the strength development of cementitious materials, but only recently their impact has been analysed on self-healing capabilities in cementitious materials. Table 1 lists various healing conditions that researchers have adopted, and the corresponding percentages of strength regained, and maximum crack widths sealed while being exposed to the various healing conditions. Yildirim *et al.* 2018 found that curing specimens in water enriched with CO_2 was able to seal crack widths up to 0.458 mm [34]. In comparison to the other healing conditions shown in Table 1, the use of CO_2-water is the most favourable strategy. This is due to the healing mechanism taking place of $CaCO_3$ formation aforementioned in Section 2, equation (1), showing the reaction between CO_2 and water, producing carbonic acid which then reacts to form hydrogen ions and bicarbonate, which subsequently reacts with calcium ions to form $CaCO_3$ precipitates. The main disadvantage of this healing strategy is that it requires samples to be cured in an artificial environment not compatible with real exposure in common structural applications. Table 1 also highlights that when using water submersion to cure cracked specimens, a maximum crack width completely healed up to 0.20 mm was observed [15,32,34,35,37]. Although greater healing abilities were observed in research conducted by Suleiman and Nehdi 2018, with crack widths of up to 0.30 mm completely sealed in the same conditions [38], such experimental variability is possibly justified by the adoption of different cement types and water to cement ratios.

When it comes to the application of self-healing concrete, the promotion of the sealing ability by methods compatible with real curing or exposure conditions for structures is of paramount importance. Therefore, researchers turned to using more realistic approaches in healing specimens to simulate outdoor environments. These include simulating external exposures by healing the specimens in wet-dry cycles with varying temperatures, as discussed in Table 1. As clearly visible in Table 1, the crack width of up to 0.05 mm was able to be completely healed [31,32]. Another interesting parameter analysed and reported in

Table 1
Review of various healing conditions and the effect it has on the performance of self-healing cementitious material.

Curing conditions	Healing Duration (days)	Strength regain (%)	Maximum crack width healed (mm)	Reference
60% RH	92	–	0	[41]
90% RH		–	0	
Wet-dry cycles (20 °C water)	–	80	0.05	[31]
Wet-dry cycles (55 °C water)	–	62	0.05	
Wet-dry cycles (20 °C water)	–	100	0.05	[32]
Wet-dry cycles (55 °C water)	–	100	0.05	
90% RH-air cycles	–	–	–	
Water submersion	–	82	0.05	
Air	–	–	–	
Water submersion	100	–	0.18	[15]
Water submersion	28	38	0.20	[37]
CO_2-Water	90	–	0.458	[34]
CO_2-Air		–	0.12	
Water		–	0.10	
Air		–	–	
90% RH	–		0	[42]
Water immersion	28		0.20	[35]
Water submersion	60		0.30	[38]

Table 1 [31,32] is represented by the influence of different water temperatures on the specimens. Yang *et al.* [31] observed that when healing at 55 °C temperature wet-dry cycle which is considered a high temperature, led to a decrease of 12% in recovered stiffness of specimens when compared to specimens healed at 21 °C wet-dry cycles [31]. Although a clear justification for this phenomenon was not provided by the authors [31,39], other studies partially confirm these findings [31,40]. The role played by the temperature is however still a subject of debate in the scientific community, where contradictory observations were reported by multiple authors [32,39,40].

A piece of work that deserves special mention analysed the influence of temperature on self-healing abilities through normalised flow rate [39]. Reinhardt and Jooss, 2003 [39] found that a temperature increases from 20 °C to 80 °C led to a significant decrease in normalised flow rate in a short amount of time. Even if a complete explanation of the underlying mechanism was not provided, the finding suggests that high temperatures can promote healing mechanisms to take place. Therefore, as can be seen, by the research reviewed in this section the influence of temperature requires more research on how it affects the autogenous mechanism and the role it plays.

3.3. *Effect of cement type*

The environmental impact of Ordinary Portland Cement (OPC) is well known and discussed in several research studies [1,43], with its increasing carbon emissions and depletion of natural resources. Therefore, over recent years a great deal of researchers have been searching for more environmentally friendly cement alternatives, which can be used as substitute for OPC. The most popular options are fly ash [44–46] and blast furnace slag [45–47], due to their capacity to bind to calcium hydroxide found in the concrete matrix. Fig. 2 illustrates the positive effect of fly ash content after 28 days of healing with 0.4 referring to the water to cement ratio. As clearly visible, increasing the fly ash content from 15 to 25% led to an increase in self-healing [45], which agrees with outcomes observed in another paper [44]. Increasing the replacement percentages of OPC using a different cement type such as fly ash with 50% and BFS with 85% leads to a decrease in crack closure [45], accompanied by a decrease in the mechanical properties of fly ash [44].

Alternative cement types, such as fly ash and BSF, provide better autogenous healing due to the delayed hydration they cause, as confirmed by Van Tittelboom *et al.,* who found that having either fly ash or blast furnace slag won't influence the amount of $CaCO_3$ produced [45]. Several researchers [45,46] found that when comparing the two cement types, blast furnace slag was the more advantageous binding material for self-healing due to its higher pH in pores, which promotes calcium carbonate crystals to develop. However, experimental investigations confirmed that specimens with crack widths < 0.50 mm could be healed regardless of the type of cement, which plays a minor role in this regard [48]. The use of various cement replacement and their influence on self-healing require intensive research to understand the extent at which they influence the healing mechanism. Alternative sustainable cement replacement is required as the current use of blast furnace slag at roughly 5–10% and fly ash approximately 3–5% of cement production as seen in Fig. 2, however, blast furnace slag and fly ash are produced from non-environmentally friendly methods. As steel production is not increasing at the same rate as cement demand and the recycling of already manufactured steel is resulting in less fly ash available [49]. As a result of the current climate emergency will push industries to look towards more sustainable cement replacement options. As seen in Fig. 3, the use of rice husk in comparison to cement is minuscule and research has proven the possible use as replacing a percentage of cement [50]. However, due to the waste material not being widely available this could be challenging because of economic viability [49]. Although, Fig. 3 shows the abundance of reserve of clay as well as it being a well distributed material, therefore can be sources globally. Therefore, shifting attention to more sustainable material with low environmental impact, in which can also promote self-healing can be seen as favourable for future research.

3.4. *Effect of crack width*

A common strategy used to observe self-healing by numerous researchers is to observe the healing of crack width [10,15,27,31,45]. Several works in the technical literature [29,32,38] show that crack size is one of the most important factors when dealing with autogenous healing. It was reported in more recent research that larger crack widths of 0.40 mm wide could not be healed, rather incomplete were observed in specimens with 0.40 mm crack width [51]. As clearly visible in Fig. 4, no crack widths healed in the literature exceed 0.30 mm. Thus, as the crack width increases self-healing by autogenous mechanism becomes less effective and complete healing may not achievable [24,27,32,51]. Despite this, even at narrower crack widths, the healing process can only

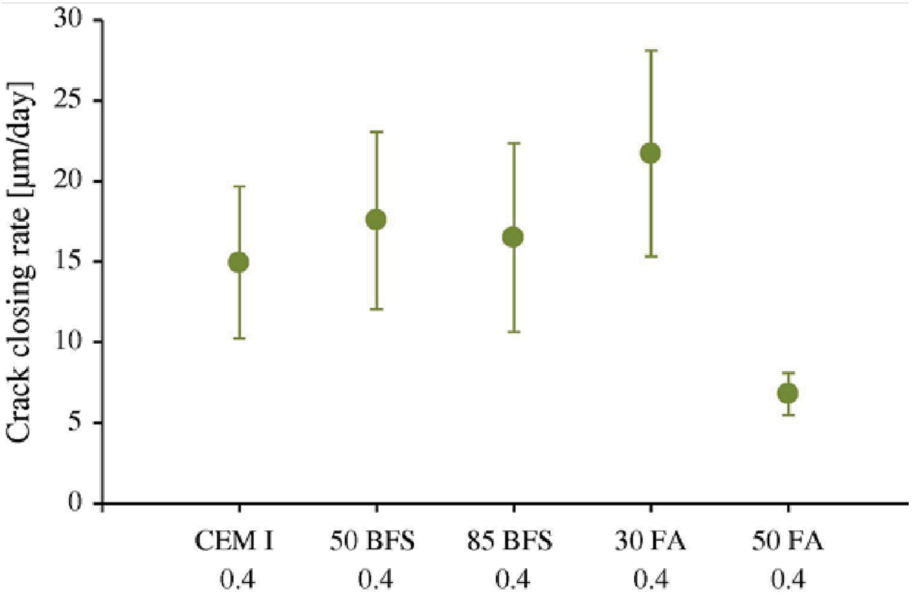


Fig. 2. Healing of cracks using various cement types and percentages [45] Copyright 2012 Elsevier Ltd.

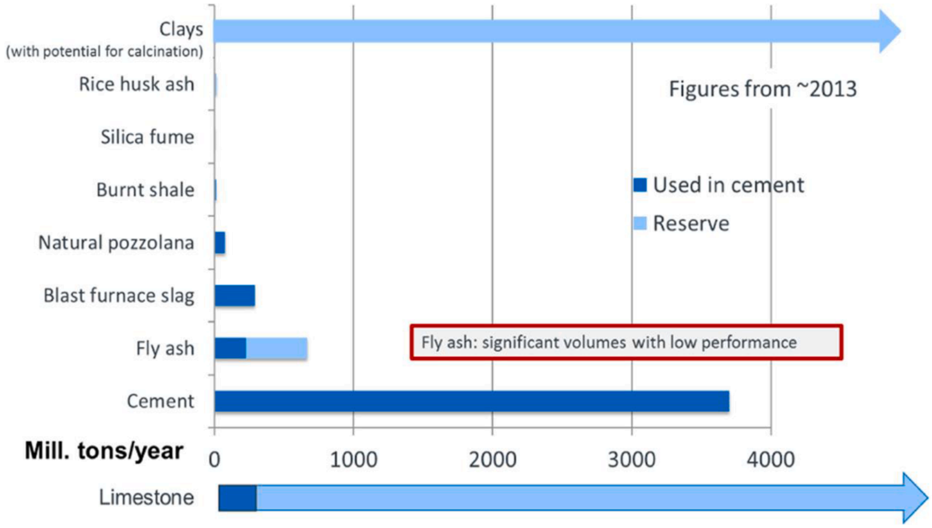


Fig. 3. Availability of binder material for use in concrete [49] Copyright 2017 Elsevier Ltd.

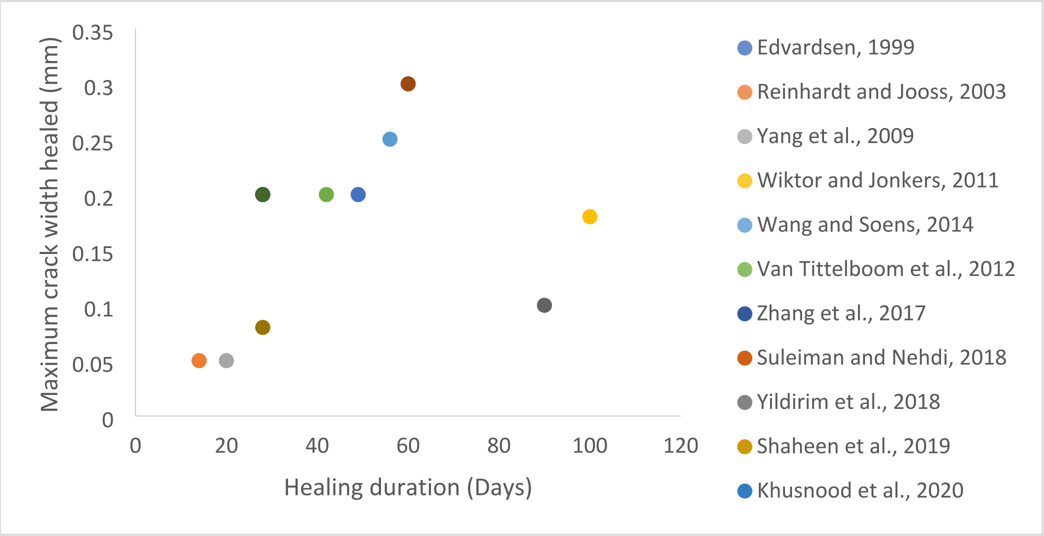


Fig. 4. Graph illustrating maximum crack width healed with time by various researchers using autogenous healing mechanism (Complied from [3,10,15,27,31,38,39,45]).

be partially exploited [32]. Indeed, crack widths within 0.138 mm-0.150 mm were reported to only partially healed [32]. Nevertheless, Fig. 5 shows quite consistent findings among different researchers work with the consensus to set the upper bound of healed crack widths at 0.20 mm.

4. Autonomous healing mechanisms

Autonomous healing is known as the engineered healing of cementitious materials due to the introduction of foreign elements, which otherwise wouldn't be originally found to promote self-healing [24].

The autonomous healing mechanism is seen as an improvement to autogenous healing because it can heal wider cracks at a faster rate due to foreign elements catalysing the reaction. Autonomous self-healing can be categorised into three categories, as shown in Fig. 5. The first is intrinsic healing which includes the use of fibres, superabsorbent polymers, and minerals to reduce the crack width and seal the crack. The second is the vascular approach that uses hollow tubes to carry various healing agents which promote self-healing. However, the most popular approach in the technical literature is represented by the adoption of capsules encapsulating the healing agents similarly to the vascular approach.

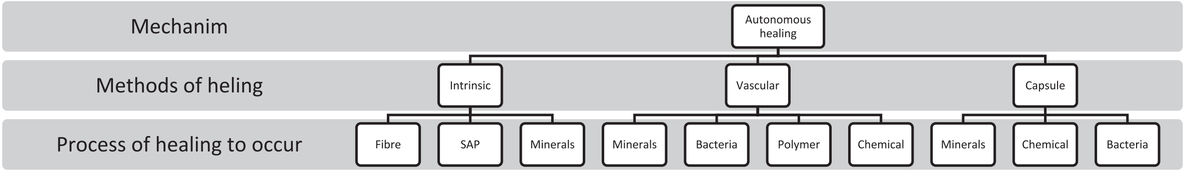


Fig. 5. Diagram illustrating the various healing mechanisms which fall under autonomous healing and the processes that healing occur.

4.1. Vascular based healing

A vascular healing system is a method used to promote autonomous healing as illustrated in Fig. 5. It involves embedding a network of hollow tubes in the concrete matrix and it is referred to as a biomimetic approach, mimicking that of the cardiovascular system seen in humans to transfer blood around the body [6]. The hollow tubes will carry the healing agents to repair the concrete when cracks form. The hollow tubes filled with healing agent are activated during the crack formation. The healing agent seeps out of the tubes into the crack through capillary action, resulting in the closure of cracks. The healing can occur through both passive and active modes of healing. The former involves a reservoir of healing agents kept outside of the concrete matrix, while the latter is a sealed hollow tube with the healing agent without any external reservoir [54]. Multiple healing cycles in the active mode of healing have also been attempted in comparison to the passive mode [55]. However, this method implies a certain degree of human interaction to refill the healing agent, therefore this might not be considered self-healing. In addition, once a crack propagates in the concrete and tubes are activated, the area where the tube broke will be the endpoint for the healing agent. This implies that the dispersion of tubes where the crack forms are of paramount importance to homogenously spread the healing agent. Davies et al., 2018, compared different methods of healing and found that when comparing bacteria and vascular healing models in large scale testing, that vascular system showed a slight increase in average crack width after 6 months of 0.13 mm, whereas bacteria and reference specimens showed a slight decrease of 0.02 mm and 0.01 mm respectively. A possible explanation to the cause of the increase has not been provided.

The vascular healing mechanism is characterised by two systems: (i) single-channel and (ii) multichannel. The former uses single-component healing agents such as epoxy or cyanoacrylate and, according to Sun, Yu and Q, 2011, the single channel systems could heal cracks up to 0.30 mm [56]. The latter system is like the single channel system, but it requires the reaction of two chemical healing agents. It requires two separate networks carrying healing agents which mix to heal the crack. One of the most used multicomponent healing agents is methacrylate [57]. The healing agent selected based on vascular self-healing system is based on low viscosity, easily penetrates cracks, chemical reactivity, wettability, and stability [6,58–60]. Joseph et al., [61], investigated the efficiency of different chemical healing agents based on several parameters, namely, cyanoacrylates resulted as a suitable healing agent due to its low viscosity, which enables them to have a greater chance of flowing into the cracks and repair properties [62–64]. However, Gardner et al., [59], states that cyanoacrylates have a self-life of approximately 1 year, in addition to rapid curing time of 1 min [60], causing it to be difficult to work with in-situ [65]. This isn't favourable for building materials as structures have an approximate service life of 50 + years, therefore selected healing agent should be stable and be useable for extended period of times due to cracks developing at any time during the service life.

However, multicomponent systems have greater stability than single component vascular system, as the multicomponent requires the mixture of chemicals. Dry and McMillan [57], report that methyl methacrylate is guaranteed to stay stable for 10 years, and experience with the use of this chemical in industry states 40 years. Another multicomponent agent is dicyclopentadiene (DCPD), in which isn't reactive to moisture and only activates ones in contact with Grubbs catalyst [66]. In addition, the adoption of a multichannel system led to the leaking of healing agents as well as the incomplete mixing of chemical healing agents [57]. Ultimately, vascular healing models prove to be an effective way of healing and distributing healing agent through the cementitious matrix, however, due to challenges presented as the stability of chemicals and implementation of vascular systems in-situ that will prove to be difficult, more research on larger scale testing would be favourable to identify the applicability of these models.

4.2. Capsule based healing

Capsule-based healing is the most promising approach of all autonomous healing methods, with great potential to heal cracks and retain mechanical properties. Capsule based healing involves confining a care material in this case being the healing agent inside a shell material, which protects the healing agent from prematurely reacting and exposed to the harsh environments found within concrete. These capsules allow the transportation of the healing agent to be embedded into the concrete matrix to provide self-healing capabilities. Capsule healing systems work by evenly distributing the capsules throughout the cementitious matrix, once cracks occur, the capsule breaks releasing the healing agent into the crack initiating the healing process. Capsules can be categorized into 2 sizes microcapsule (<1mm) and macrocapsules (greater than1mm). In this section of the review macrocapsule and microcapsules will be covered with nanosized capsules covered later in the review. The varying sizes in capsules can have an influence on mechanical properties as well as healing probability, as the larger the capsule the less can be embedded into the cementitious material [52]. However, the loading capacity of with macro-sized capsules is greater, possibly providing better healing. In the technical literature, various different capsules have been tested for instance glass [19], lightweight aggregate (LWA) [18,53], Polyurethane (PU) [54,55] urea–formaldehyde (UF) [17], poly(lactic acid) (PLA) [56] and polystyrene (PS) [56]. Capsules to be considered for self-healing applications are required to i) survive mixing of cementitious material and ii) rupture once cracks occur. Therefore, it has be flexible enough to survive mixing of cementitious material due to the mechanical stress that is applied to the capsule during the mixing phase, as well as be flexible enough to rupture when cracks occur to enable the healing agent to flow out into the crack.

The use of macro-sized capsules have been extensively studied with Hilloulin et al., [56] studying the use of PLA, PS and PMMA/N-BMA with capsule diameters of approximately 1.9–52 mm, 6–6.8 mm and 7.2 mm respectively and Formia et al., [57], studying cementitious hollow tubes with diameters of 2 mm and 7.5 mm. Hilloulin et al., [56], found that the use of heat to make the capsule material flexible had positive influence on the survival of capsule during the mixing of concrete, in which found that 80%, 90–100% and 100% of PMMA/N-BMA, PS and PLA respectively were able to survive the mixing. Once embedded in the concrete the previously mentioned materials will have a brittle nature once cooled and rupture when cracks occur. The most significant parameters for capsule-based material are its geometry, thickness, and diameter. Its geometry, whether its spherical or cylindrical, plays a crucial role not only in mechanical properties but also survivability of the capsules. Another study developed an analytic model and found that the efficiency of self-healing is linearly proportional to the aspect ratio, however, there were lack of consideration for the survivability of the capsules [52]. Sinha et al., [58], through experimental evidence found that increasing the aspect ratio from 1 to 1.5 observed a decrease of survivability of 30%. Therefore, both [52] and Sinha et al., [58], show the importance of considering the design of capsules to create a suit capsule for self-healing application.

Although capsule design is of utmost importance for successful transportation and effective healing of healing agents. However, healing agents also play a significant role in the healing efficiency of cracks, hence Table 2 shows the various healing agents used by researchers in literature. In previous literature [17–19,54,55], sodium silicate were popular healing agent options as seen in Table 2. Gilford III *et al.* studied the effectiveness of various parameters, namely shell thickness, diameter size of capsules, temperature, and rate of agitation of microcapsules and their influence on self-healing [17]. According to the experimental findings, increasing the healing agent led to an increase in the healing rate, in detail, using 5% led to an 11% increase in modulus of elasticity [17]. Whereas doubling the concentration of sodium silicate from 2.5% to 5% was reported to lead to an increase in the healed depth of the crack in two weeks as well as recovery of 20–26% of its original strength [54].

Table 2
Review of various capsule-based healing.

Shell material	Healing agent	Concentration	Performance	Reference
Urea-Formaldehyde	Sodium silicate	5%	• Increased the modulus of elasticity by 11% after cracking.	[17]
Urea-Formaldehyde	DCPD	0.25%	• Increased the modulus of elasticity by 30% after cracking.	[17]
LWA	Sodium silicate	–	• Maximum crack width healed 0.135 mm • Recovered 80% of original flexural strength	[18]
Glass	Sodium silicate	–	• Maximum crack width healed 0.20 mm • Recovered 20% of loading	[19]
Polyvinyl alcohol (PVA)	Granulated calcium sulphoaluminate (CSA)	10% by mass of cement	• Maximum crack width healed 0.182 mm • Decreased water permeability between 80 and 90%	[59]
Dirctly applied	Crystalline additives (CA) and CSA	10 %CSA And 5% CA	• Maximum crack width healed 0.40 mm 92% decrease in water passing rate after 56 days.	[20]
Double walled PU/UF	Sodium silicate	2.50% 5%	• Healing efficiency significantly increased by 23% and 35% for concentration 2.5% and 5% respectively.	[54]
Polyurethane	Sodium silicate	–	• Recovered 20–26% flexural strength after being cracked.	[55]
Glass	Polyurethane	–	• 67% of the specimens with crack width of 0.10 mm, almost 100% healed. 50% of specimens with 0.30 mm cracks showed comparable chloride penetration to cracked specimens.	[75]
Cementitious tubular capsules	Polyurethane	0.3–0.9 mL	• Healing rate of 35.9%-46.5%. Regain of 50% and 82% of flexural strength after 1st and 2nd reloading respectively.	[76]

In another piece of literature, the use of sodium silicate in LWA, enabled the recovery of 80% of flexural strength, accompanied by 0.135 mm crack width fully healed [18]. However, Table 2 indicates other researchers observing lower rates of strength recovery of approximately 20% [19], and 26% with concentrations of 5% sodium silicate [54]. This suggests that the effectiveness of self-healing can vary even while using the same healing agent, implying immobilization materials may also impact strength recovery when using it to measure self-healing.

Table 2 shows that urea–formaldehyde shells aren't as effective as lightweight aggregate when it comes to evaluating the regain of mechanical properties. This is possibly due to the interfacial bonding between the matrix and the shell material, causing the material to be weaker. According to Gilford *et al.,* [19], the use of dicyclopentadiene (DCPD) as a chemical healing agent at a 0.25% concentration led to a 30% increase in modulus of elasticity, this is almost 3 times that of when 5% concentration of sodium silicate was used at a concentration 20 times greater [17]. A promising healing agent used by Lee and Ryou [59] was calcium sulpho aluminate (CSA) granules, which led to 100% healing of cracks up to 0.182 mm wide within 16 days, whereas reference specimens could only heal 20–80% within 28 days [60]. Sisomphon, Copuroglu and Koenders also mixed CSA with crystalline additive and observed healing of a significant crack width of up to 0.40 mm [20], which is greater than what was obtained by Lee and Ryou [59] by using CSA only. The use of CSA and crystalline admixture appears to represent a more efficient method compared to the use of sodium silicate shown in Table 2.

In addition, polyurethane (PU) is often used as a healing agent, as seen in Table 2 with researchers reporting success in both durability and regain of mechanical properties. Maes, Van Tittelboom and De Belie, [61] studied the healing capabilities of polyurethane in macrocapsules as a potential to resist chloride environments from entering cracks. It was found that 67% of the specimens with PU resulted in almost fully regaining resistance to chloride penetration at a crack width of 0.10 mm. Whereas once the crack width is increased from 0.10 mm to 0.33 mm only 33% of specimens showed no chloride ion penetration until the glass tube, with 50% observing similar results as crack specimens (no resistance) [61]. Anglani et al., [62], observed more promising results when using PU as a healing agent. The author reported a healing rate of 35.9%-46.5%, in comparison to the control specimen only showing 0.1% healing. It was also found that the use of larger diameter microcapsule of 7.5 mm with epoxy external coating led to 50% regain flexural load capacity upon first reloading and 82% regain after the second reloading. The difference in regain strength was attributed to the hardening of the PU sealing both the crack and the microcapsule. In which when the second unloading occurred there were unpolymerized PU which was able to be used in the second time of healing hence the increase in regained flexural strength [62]. Ultimately, this indicates the potential of using macrocapsules with PU to improve mechanical properties after healing and potential repeat healing due to the properties of the healing agent which can protect or seal unreacted healing agents inside the capsule. Overall, the use of capsules is an effective way of protecting the healing agent from prematurely activating during transportation into the cementitious structure [63] and in the case of bacteria protecting from mechanical stresses as well [15], which will be discussed in this review.

5. Bacteria healing

Bacteria healing has been extremely popular as chemical and mineral healing agents have an environmental complication and their healing results are not very promising. Researchers have now turned their attention to the use of microbially induced calcium carbonate precipitate as an alternative method [4,10,14,16], and have found that the introduction of bacteria to concrete improves its permeability and durability, and such increasing the life span of the cementitious material and improves its mechanical properties due to densification of the microstructure[4,10,14,16]. Bacteria are selected for self-healing concrete if they meet several criteria, namely, alkaliphilic to survive pH conditions around 8–13, which is found in concrete [4,11,14]. Bacteria produced as spores are more likely to survive both mechanical stresses and harsh conditions found in concrete [36] and they can lay dormant for up to 200 years [64], which is ideal for the longevity of structural materials. Bacteria have to have oxygen tolerance [4], as oxygen isn't guaranteed within the material matrix. The selected strains of bacteria must be able to function in both aerobic and anaerobic conditions [4,11,14,36]. The most used bacteria in research which meets the criteria set above is *Bacillus*, which is a rod-shaped bacterium [14,36]. Table 3 shows several different bacteria which have been used in works of literature, they show the various healing mechanisms that can occur to develop healing products which will be further explained in a subsequent section.

5.1. Healing mechanisms

There are several bacteria-based healing mechanisms to produce $CaCO_3$ precipitates to heal cracks. The most important are (i) enzymatic ureolysis, (ii) metabolic conversion of organic acid, and (iii) nitrate reduction. Enzymatic ureolysis and metabolic conversion of organic precursor are two commonly researched mechanisms. The nitrate reduction mechanism occurs when oxygen is limited while, the

Table 3
Review of various bacteria used in literature along with how they were applied and the healing mechanism.

Healing mechanism	Bacteria strains	Method of application	
Metabolic conversion of organic precursor	B. Pseudofirm DSM 8715	Direct	[4]
	B.Cohnii DSM 6307		
	B. Subtilis	Direct	[35]
	B. Subtilis	Immobilized	
	B. Cohnii	Direct	[3]
	B. Cohnii	Immobilized	
	B. Pasteurii	Immobilized	[65]
	B. subtilis	Immobilized	[11]
	B. Subtilis	Direct	[66]
	B. Alkalinitriicus	Immobilize	[15]
Ureolytic Hydrolysis	B. subtilis	Direct	[67]
	B.halodurans	Direct	
	B.licheniformis	Direct	
	B. Sphaericus	Immobilized	[10]
	S. Pasteurii	Direct	[68]
	Lysinibacilluss Sphaericus	Direct	
	S. Pasteurii	Immobilized	[69]
	B. megaterium	Direct	[70]
	B. subtilis	Direct	
	B. Sphaericus	Immobilized	[71]
	CERUP	Immobilized	
	S. Pasteurii	Immobilized	[72]
	B. Sphaericus	Immobilized	[73]
	B. Pasteurii	Direct	[74]
	B. Sphaericus	Immobilized	[16]
	B. Sphaericus	Immobilized	[8]
Nitrate Reducing	D. Nitroreducens	Immobilized	[71]
	ACDC	Immobilized	
	S. Pasteurii	Immobilized	[75]
Unknown	Shewanella	Direct	[13,76]

metabolic conversion reaction occurs due to bacteria acting as a catalyst, converting the organic precursor of choice to $CaCO_3$ to seal the crack. Jonkers in [4] found that a suitable precursor should be selected for this reaction as it could influence the strength and setting time of concrete. The following reaction shows the metabolic conversion of the organic precursor (calcium lactate) to form $CaCO_3$ for healing [14,15]:

$$CaC_6H_{10}O_6 + 6O_2 \rightarrow CaCO_3 + 5CO_2 + 5H_2O \tag{4}$$

Eq. (4) illustrates the reaction producing $5CO_2$ molecules, which can react with the $Ca(OH)_2$ found within concrete producing 5 more $CaCO_3$ molecules. This reaction is 6 times more effective than autogenous healing, hence why it can close greater crack widths [77]. The next mechanism is enzymatic ureolysis it works by producing urease enzyme by bacteria which accelerates the hydrolysis of urea to one mole of ammonium and carbonate. This reaction causes an increase in pH and carbonate concentration. The reaction is as follows [78]:

$$CO(NH_2)_2 + H_2O \rightarrow NH_2COOH + NH_3 \tag{5}$$

$$NH_2COOH + H_2O \rightarrow NH_3 + H_2CO_3 \tag{6}$$

$$H_2CO_3 \leftrightarrow HCO_3^- + H^+ \tag{7}$$

$$2NH_3 + 2H_2O \leftrightarrow 2NH_4^+ + 2OH^- \tag{8}$$

$$HCO_3^- + H^+ + 2NH_4^+ + 2OH^- \leftrightarrow CO_3^{2-} + 2NH_4^+ + 2H_2O \tag{9}$$

$$Ca^{2+} + Cell \rightarrow Cell - Ca^{2+} \tag{10}$$

$$Cell - Ca^{2+} \rightarrow Cell - CaCO_3(\downarrow) \tag{11}$$

The bacteria cell wall serves as a nucleation site due to its negative charge which attracts positive cations from the concrete matrix, e.g. Ca^{2+}. Ca^{2+} deposits on the cell surface and reacts with CO_3^{2-} and it forms the $CaCO_3$ precipitate useful for sealing cracks [78]. The last mechanism covered in this section is nitrate reduction which can only occur with bacteria able to respire NO_3^-. The pathway for this reaction is as follows [79]:

$$Ca(HCOO)_2 + 0.4Ca(NO_3)_2 \rightarrow 1.4CaCO_3 + 0.4N_2 + 0.6CO_2 + H_2O \tag{12}$$

$$2HCOO^- + 2NO_3^- + 2H^+ \rightarrow 2CO_2 + 2H_2O + 2NO_2^- \tag{13}$$

Eq. (12) shows the calcium formate reacting with the organic precursor, in this case, calcium nitrate is used to produce $CaCO_3$, and the by-products are nitrogen, carbon dioxide, and water. The CO_2 produced can then react with $Ca(OH)_2$ found within the matrix to form $CaCO_3$ precipitates. As visible in equation (13), the formate ion reacts with the nitrate and hydrogen ions to produce nitrate groups (NO_2^-) which is known to be a corrosion inhibitor [79]. In the following sections, some of the most important parameters affecting the mechanisms discussed above will be critically reviewed.

5.2. *Factors influencing bacteria healing mechanisms*

In this section, the factors influencing the bacteria healing mechanisms will be reviewed focusing on those factors which can influence the reactions to precipitate calcium carbonate. The following factors can have positive or negative influences on how effective bacteria healing can be.

5.2.1. *Effect of bacteria type*

Fig. 6 illustrates the influence that various bacteria have on the mechanical property of cementitious materials. Ramachandran, Ramakrishnan and Bang introduced B. *Pasteurii,* which led to an 18% increase in compressive strength [74]. Ghosh *et al.* used Shewanella, which exhibited the greatest increase in compressive strength of 25% [13]. This increase in compressive strength due to the addition of bacteria can be attributed to the production of $CaCO_3$ crystals being formed by the reaction seen in Eqs. (5)–(11). The $CaCO_3$ produced by the bacteria fill the pores in cementitious material. Therefore, making the cementitious microstructure more compact and denser, improving its compressive strength. However, Fig. 6 also shows that the introduction of B. *Pseudofirmus* and B. *Sphearicus is often associated with a* decrease in the compressive strength of 10% [4] and 35% [80], respectively. This is probably justifiable by the interaction of various other factors, such as bacteria concentration and curing conditions, which will be discussed in more detail in sections to follow.

5.2.2. *Effect of pH*

Cementitious materials are known to present unfavourable conditions affecting the growth and survival of bacteria. Several researchers studied the influence that various pH conditions have on the size, texture, and shape of the $CaCO_3$ formed from healing [81]. Multiple researchers conducting pH-based experiments to examine the influence it has on bacteria observed that increases in the pH led to a decrease in bacterial activity [16,81,82]. Therefore, several studies have agreed that the urease activity is dependent on pH when healing occurs through the enzymatic ureolysis pathway [81-83]. Wang et al. [16] studied the influences of pH on non-protected bacteria to identify its effects on the bacteria activity and it was found that increasing the pH from 7 to 12.5 led to a significant decrease in ureolytic activity compared to encapsulated bacteria. Similar findings were obtained in studies conducted by Wu et al., [23], Stocks-Fischer et al. [80] and Lee [81], supporting the argument that an increase of pH beyond 8 was associated with a decrease in the urease activity [82,84], and as a result reduction in calcite produced (Fig. 7). This ultimately suggests that bacteria require encapsulation and protection to stay viable in concretes characterized

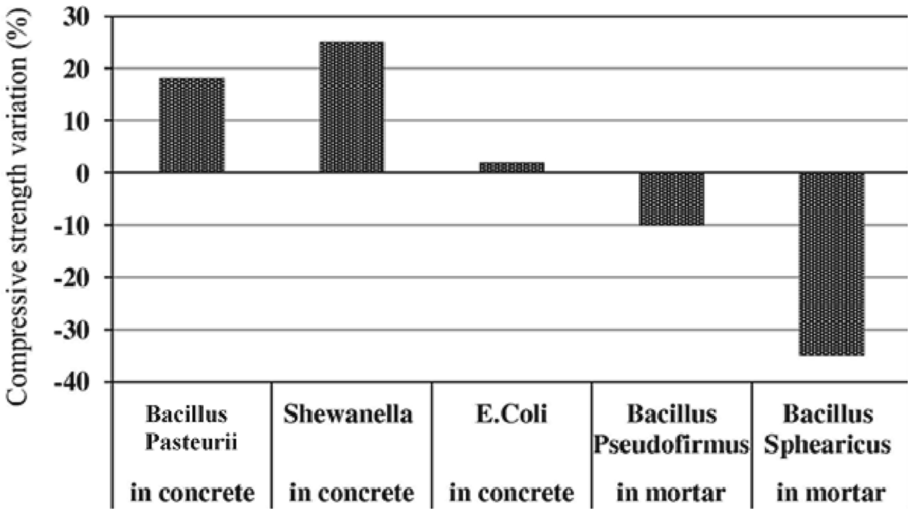


Fig. 6. The influence of various types of bacteria has on concrete and mortar specimens [14] Copyright 2015 Elsevier Ltd.

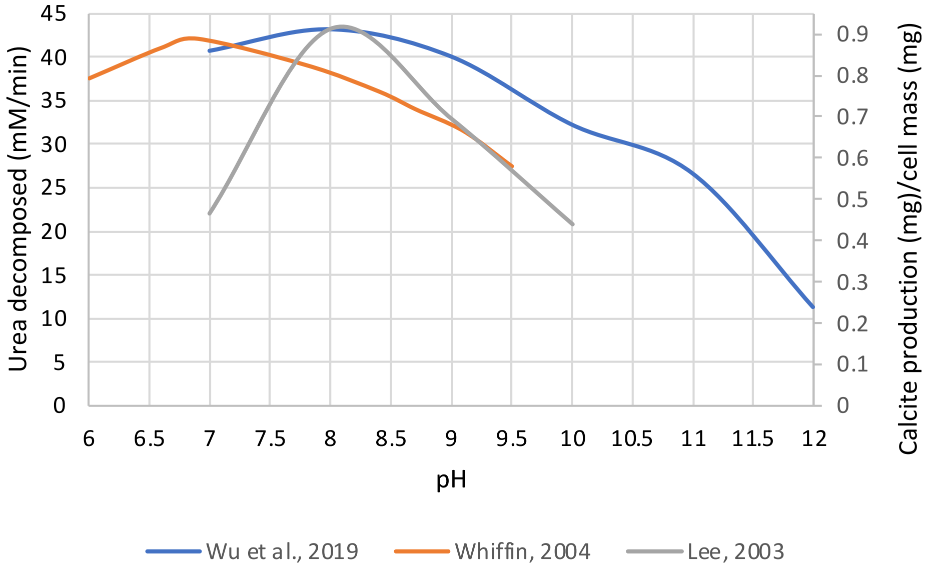


Fig. 7. Influence of pH on ureolytic activity in terms of urea decomposed and calcite production (Orange and blue lines represent pH influence on urea decomposed, grey line represent pH influence on calcite production). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

by a pH higher than 8.

5.2.3. *Effect of temperature*

Temperature is another factor that influences the healing mechanism with bacteria healing agents. As depicted in Fig. 8, temperature influences urease activity. Indeed, bacteria contain the enzyme urease which catalyses the hydrolysis of urea, and this process is known to be dependent on temperature. Previous research [85] demonstrated that when temperature is increased, bacteria activity also increased up to an optimum temperature of approximately 69 °C [23]. Whiffin [85] and Wu et al. [23] studied the influence of temperature on bacterial activity by observing urea decomposition. They discovered that increasing the temperature from 10 °C to 40 °C also increased bacterial activity (Fig. 8) [23,85]. However, the effectiveness in different temperatures is dependent on the type of bacteria selected, as some, such as psychrophiles, can grow effectively in rather cold climates (−20 °C to 20 °C), whereas hyperthermophiles prefer warmer temperatures (88 °C −106 °C) [86].

5.2.4. *Effect of nutrients*

Nutrients are an essential part of self-healing cementitious materials. They comprise microorganisms which are food for bacteria to grow and reproduce to be able to produce $CaCO_3$ precipitates required for healing. Fig. 9 illustrates the different morphologies achieved by using various calcium sources to produce $CaCO_3$, calcium nitrate and calcium lactate were seen to produce spherical and rhombohedral shaped $CaCO_3$ with small particle size [87]. Both calcium sources in Fig. 9 produced similar SEM morphology of $CaCO_3$. However, Gorospe found that calcium chloride can provide rhombohedral crystals with smooth surfaces and shape rigid edges whereas calcium acetate is characterized by a lettuce texture [83]. Gorospe et al. [83] conducted a microscopic investigation experiment using SEM to study the morphologies of crystals and found that calcium lactate produced the largest crystals with calcium acetate coming second [81].

The selection of a calcium source (precursors) to produce $CaCO_3$ in concrete is crucial. Fig. 10 depicts the effects of various precursors on the compressive strength of concrete specimens during the laboratory investigation conducted by Jonkers *et al.* [4]. It was found that in [4] the

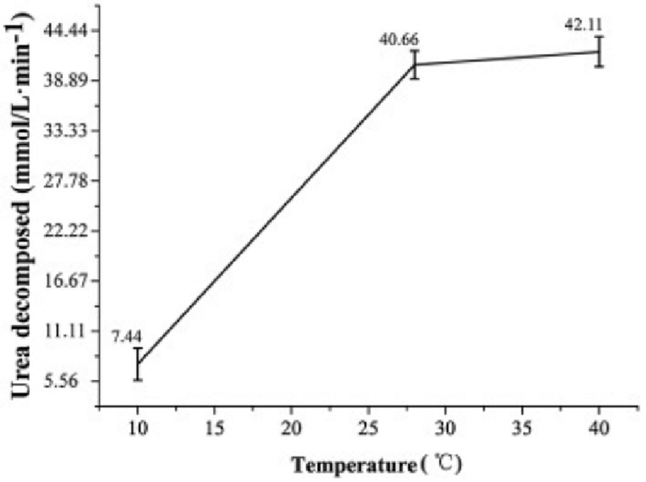


Fig. 8. Effect of temperature on Urea decomposed [23] Copyright 2019 Elsevier Ltd.

addition of 1% calcium lactate led to a slight increase of 20.75% in the compressive strength when compared to the control whereas other precursors used such as peptone, which showed the biggest detrimental influence on concrete compressive strength (Fig. 10a). Khaliq and Ehsan [14] and Khushnood *et al.* [35] opted for the use of calcium lactate due to its non-destructive influence on concrete. However, in an experiment conducted by Paine [88], which saw the effects of different concentrations of calcium lactate, it was found that when calcium lactate concentration surpasses 1%, compressive strength decreased. Contradicting findings were also reported that [89] the compressive strength and flexural strength increased by approximately 18% and 12% respectively, compared to the control specimen when using calcium acetate as a precursor as seen in Fig. 10b. Whereas in Fig. 10a it can be seen that [4], calcium acetate caused a decrease in compressive strength. Such outcomes could be due to different concentrations of the precursors used, but more research needs to be conducted on the number of other calcium precursors and cheaper alternative precursors to evaluate their influence on the concrete properties.

5.2.5. *Urea and Ca^{2+} concentrations*

The possible reaction that bacteria take part in to produce $CaCO_3$ is by utilizing the urease enzyme. It is understood that urea is hydrolysed to 1 mol of ammonium and carbonate, and such has a crucial part to play and influence the ureolytic activity of bacteria, hence influencing its ability to precipitate calcium carbonate. Several researchers [23,90] found that the increase in urea concentration led to a decrease in bacterial activity and this can be seen in Fig. 11a. In a study conducted by

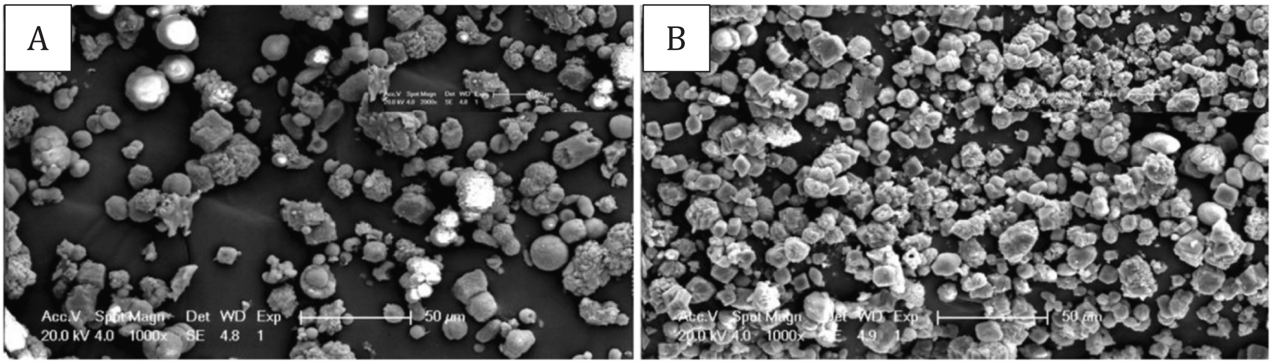
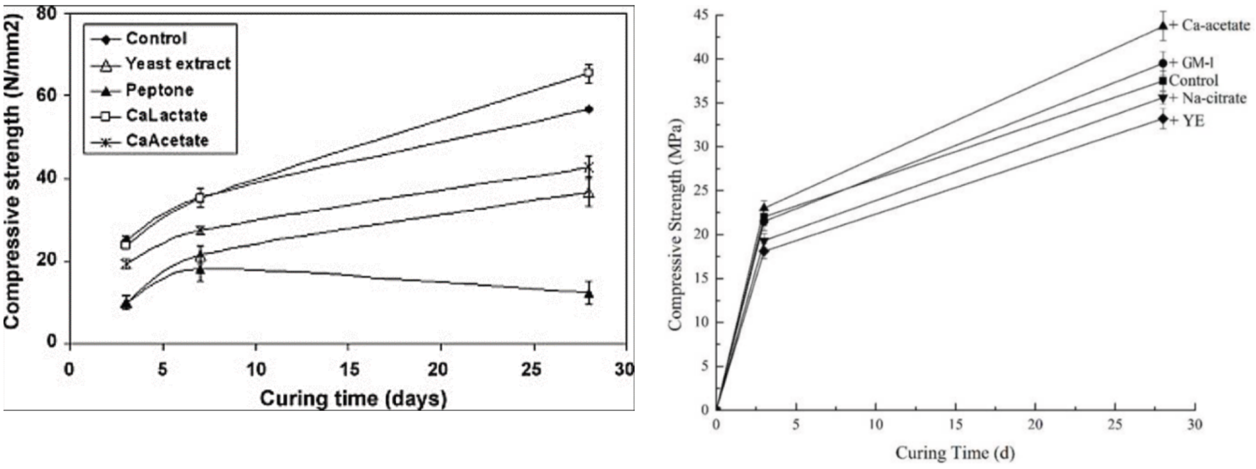


Fig. 9. SEM images of the morphology of calcite crystals when using various calcium sources: a) calcium nitrate, and b) calcium lactate [87] Copyright 2020 Elsevier B.V.



a

b

Fig. 10. Influence of various precursors used in MICP on the compressive strength of the specimen with respect to curing time a) [4] Copyright 2009 Elsevier B.V. b) Reproduced from [89] Copyright 2019 by MDPI AG.

Wang *et al.* [90], they were able to see that when increasing the concentration of urea from 90 g/L to 180 g/L at a cell concentration of 10^5 cells/mL, the amount of the decomposed urea decreased. As previously mentioned, this cell concentration is to provide the optimum mechanical properties. Wang *et al.* [90] were not able to explain why excess urea causes the decrease in urea decomposition, this area requires further research. It was reported that [23,90,91] an optimum urea concentration dependent on the concentration of cells is 0.60–0.80 M [23,90,91], which has been shown by laboratory evidence. However, Wang *et al.* [92] found that higher bacteria cells allow for a higher concentration of urea without hindering urea decomposition, whereas when the bacteria cell concentration decreases to 10^7 cells/mL, it is able to see that when urea concentration passes 1.5 M, the decomposed urea decreased. Wang *et al.* [90] were also in agreement with that excessive amount of urea had a negative influence on ureolytic activity. A likely explanation for this is that bacteria have a set amount of urea, which can be hydrolyse, as a result, once its limit is surpassed, the efficiency decreases [92].

Another important component in self-healing to take place is the concentration of calcium ions (Ca^{2+}). As aforementioned, the Ca^{2+} is used in the reaction to produce calcium carbonate on the bacteria cell wall. It was seen by Okwadha and Li [91] that by increasing the Ca^{2+} concentration from 0.025 M to 0.25 M, there was over 100% increase in the $CaCO_3$ produced and this was irrespective of urea concentration. It can also be seen that lower concentrations of Ca^{2+} cause more urea decomposition compared to higher concentrations such as 1.2–1.5 M (Fig. 11b) [23]. Wang *et al.,* [90] have also reported that an increase in Ca^{2+} concentration greater than 0.5 M caused a decrease in urea decomposition and concluded that 0.5 M is the optimum concentration for Ca^{2+}. However, several researchers opted for the use of 1 or 2 concentrations, which is a very small sample and insufficient of being able to see the true influence that the concentration of Ca^{2+can} have. Whereas Wu et al. [23] was able to conduct the research with multiple different concentrations (Fig. 11b) and able to see a negative trend, when Ca^{2+} concentration increases and urea decomposition decreases. This is due to that an excessive amount of Ca^{2+} can become toxic to the bacteria, as the bacteria only require a limited amount, as well as only 0.02 M of Ca^{2+} could be present in the crack zone due to calcium hydroxide dissolved [92]. This is why bacteria selection is also crucial to achieve a high tolerance to Ca^{2+}.

5.2.6. *Effect of cell concentration*

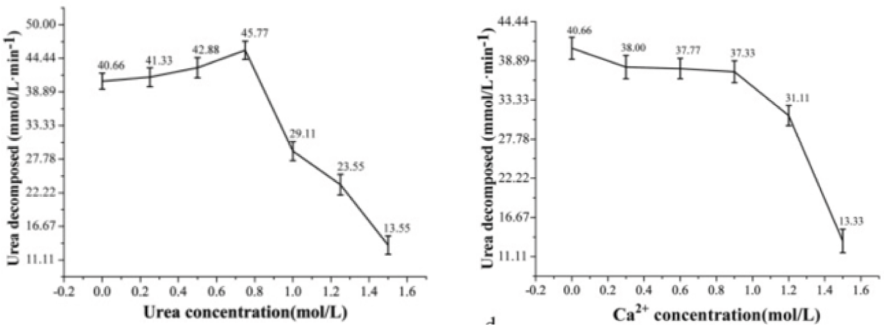
Cell concentration of bacteria is a significant factor influencing the self-healing capabilities of cementitious materials. Table 4 summarizes some of the most important research work focused on investigating the influence of cell concentration on the self-healing capabilities of concrete in terms of its mechanical properties and strength regain. Ghosh *et al.,* [13] used Shewanella bacteria at concentrations of 10^3, 10^5, and 10^7 cells/mL, and observed an increase in compressive strength from cell concentration of 10^3 to 10^5 cells/mL after 28 days of curing. Such an increase in compressive strength suggests that internal healing is taking place due to the bacteria introduced into the concrete matrix. When the cell concentration reached 10^7 cells/mL, a decrease in strength was observed by researchers [13,93,94]. However, Ramachandran *et al.,* obtained contradictory findings on cell concentrations influence, as an increase in strength was observed with a specimen with a cell concentration of 7.6×10^3 cells/mL, and no influence in strength was observed for a cell concentration of 7.6×10^7 cells/mL [74]. A likely explanation for this variation could be due to the type of bacteria being used, which also has a significant influence on healing capabilities.

Several previous studies [13,93-95] agreed that the optimum cell concentration to use for self-healing also characterized by avoiding a negative impact on the strength of the material would be 10^5 cells/mL regardless of the bacteria type [13,93-95]. A likely explanation for the increase in strength is due to bacterial activity, which subsequently increases $CaCO_3$ precipitate production. As the bacteria cell acts as a nucleation site for the reaction of the healing product to be formed, the increase in available nucleation sites leads to more $CaCO_3$ being produced. An excessive amount of $CaCO_3$ may block pathways for the bacteria to utilise the nutrients. Thus, a decrease in strength is observed. This is usually associated with increases in the cell concentration beyond 10^5 cell/mL.

5.2.7. *Effect of processing*

The application of bacteria has a significant influence on the self-healing of cementitious materials. The way the bacteria are introduced into the concrete matrix determines the effectiveness of self-healing [4]. The direct application of bacteria into the concrete matrix led to a decrease in the amount of $CaCO_3$ produced [4]. Jonkers *et al.* justified such findings by a decrease in the viability of the bacteria as a result of the mechanical stresses due to the mixing of the cementitious material and the high alkaline pH [4]. In the technical literature, several scholars [4,10] highlighted the importance of finding a way to protect the bacteria from such harsh conditions. To improve the bacteria viability, researchers considered using a novel approach to encapsulate bacteria using porous materials, including lightweight aggregate [15], graphite nanoparticles (GNP) [14], iron oxide nano/microparticles (IONP) [36], hydrogel [96], diatomaceous earth [16], expanded clay [3,10,15], expanded perlite [3], and natural fibres [97]. Multiple researcher's work [3,14,35,36], confirmed that the adoption of this strategy led to an increase in crack widths being healed, regardless of the type of encapsulating porous material used [3,14,35,36].

Jonkers *et al.* found that as the number of curing days increased, as time progressed from day 9 to day 22 of curing, the number of viable cells approximately decreased 83.33% [4], which suggests a significant number of bacteria cells were unable to survive. However, Wang *et al.* [96] provided a possible explanation for the decrease in viability, which



a b

Fig. 11. Influence of various (a) urea concentrations and (b) calcium ion concentrations on the decomposition of urea [23] Copyright 2019 Elsevier Ltd.

Table 4
Review of the influence of bacteria cell concentration on self-healing.

Type of Bacteria	Concentration (cells/mL)	Days healing	Performance of strength	Reference
B. Sphaericus	10^3 10^5 10^7	21	• Strength increase of 20.8% • Strength increase of 37.5% • Strength increase of 33.3%	[94]
B. Pseudofirmus	6×10^8	28	• Compressive strength decreases by 10%	[4]
B. Pasteurii	7.6×10^3 7.6×10^5 7.6×10^7	28	• Strength increase of 18% • No strength change • No strength change	[74]
Shewanella	10^3 10^5 10^7	28	• Strength increase of 9.8% • Strength increase of 25.3% • Strength increase of 11.1%	[13]
S. Pasteurii	10^3 10^5 10^7	28	• Strength increase of 4.2% • Strength increase of 16.7% • Strength increase of 8.3%	[93]
B. Aerius	10^5	28	• Strength increase of 72.7%	[95]

occurs as the number of curing days increases, and it was considered that this is due to bacteria cells ranging from 1 to 3 μm and it is estimated for pore sizes in a cement-based matrix to be 0.5 μm as the microstructure of the cementitious material becomes denser, leading for pores to get smaller [96]. Several researchers[14,35,36] also observed that as the number of days of curing increased, the specimen's microstructures become denser. Also, as the hydration process continued, the microstructure developed and underwent densification, and the pore sizes decreased, which caused uniaxial loading on the bacteria, which in turn caused them to be crushed [96]. This then renders the bacteria to be useless and unable to induce $CaCO_3$ precipitate, therefore decreasing the specimen's self-healing capability.

5.2.8. *Effect of pre-cracking age*

Pre-cracking age has a significant influence on the healing mechanism in cementitious materials (Table 5). A trend can be observed that as the pre-cracking age of specimens increases, the healing efficiency and recovery of strength gradually decrease. This trend is also seen in the autogenous mechanism (see section 3.1), as the age of the specimen increases, the healing ability reduces. It was reported that at 3-days of pre-cracking, wider cracks could be healed compared to specimens pre-cracked at 28-days [14]. A compilation of previous findings showed that specimens that were pre-cracked at 3-days healed cracks 1.2 times wider than specimens pre-cracked at 28-days (Table 5) [14]. Rauf *et al.* [97] observed an estimated 1.8 times healing when comparing young and mature specimens. Conversely, to mature specimens, younger specimens are characterized by higher availability of unreacted cement particles which can promote the hydration process and help close wider cracks [33]. Another possible explanation is linked to the long curing period in mature specimens. A longer curing period implies fewer pore spaces available within the matrix, which can cause premature crushing of the capsules, making the bacteria useless and reducing bacterial activity. However, different findings were also reported [35] where laboratory evidence suggests an increase in the pre-cracking age, led to wider crack width to be healed, and this was explained that the increase was due to uniform distribution of bacteria accompanied by better protection of the carrier material.

5.2.9. *Effect of healing conditions*

Healing conditions is very important and can influence both the healing capabilities and the strength development in concretes. Table 6 shows a summary of various healing conditions used by different researchers and the corresponding maximum crack healed. According to the technical literature [10,15,99], the presence of water is vital for self-healing to take place. Indeed, microorganisms (spores) are not able to grow and undergo binary fission without the presence of water and nutrients. This is also confirmed by [10] laboratory evidence, which observed that when using 95% relative humidity no changes in crack size after 56 days of healing were detected. In addition, regardless of the bacteria healing agent or autogenous healing, the presence of water is crucial to trigger both healing mechanisms. Several researchers used water immersion as a healing option to promote self-healing in specimens [10,15,79,94,100]. Table 6 suggests that water immersion is an efficient way to promote healing of crack widths between 0.45 mm and 0.97 mm. Some variability in experimental investigations using the

Table 6
Review of healing conditions utilised in the literature.

Curing conditions	Days of healing	Maximum cracks healed (mm)	Reference
95 %RH	56	0	[10]
Water immersion		0.85–0.97	
Medium immersion		0.33–0.40	
Wd-water		0.54–0.60	
Wd-medium		0.28–0.29	
Water immersion	100	0.46	[15]
Water immersion	56	0.48	[79]
Water immersion	21	–	[94]
Water immersion	20	0.2	[100]

Table 5
Review of literature on the influence of Pre-cracking.

Type of Capsule	Pre-crack (days)	Healing (days)	Strength recovery (%)	Maximum cracks healed (mm)	References
LWA	3 14 28	28	–	0.63 0.59 0.51	[14]
Iron oxide nano/microparticles	3 28	28	60% 46%	–	[36]
No encapsulation	3 28	28	87% 47%	–	[98]
Recycled coarse aggregate	3 28	28	76% 73%	0.44 0.70	[35]
Natural fibres	7 28	28	–	0.70 0.40	[97]

same healing condition is possibly due to factors, such as bacteria concentration, type of bacteria, application, and pre-cracking age. An increase in healing duration could also lead to a decrease in crack width [14,35]. Wang *et al.,* [10] observed that the use of water immersion outperformed the healing condition of medium immersion with the latter being made of calcium nitrate and urea to accelerate the healing process. Although wet-dry cycles (Wd) in water (Wd-water) or medium (Wd-medium) didn't perform to the same standards as water immersion (Table 6), it is the only healing method, which mimics that of the environment the cementitious elements will encounter, however, that being said if applications for water retaining structures are required it shows the use of bacteria to be effective for example for dams, piers and sea wall.

5.2.10. Effect of crack width

Crack width is the most important parameter for evaluating self-healing mechanisms, as it allows for the quick identification of whether healing is actually taking place. Therefore, it is common practice to use it as an indicator of healing, alongside other methods of characterising healing [3,10,14,15,35,36,96,98]. Although crack width is very important, it is not independent of other factors, such as curing conditions and pre-cracking age. Fig. 12 illustrates a summary of the various immobilizing materials and bacteria and their influence on crack width. It was reported that [3,10,15] the use of expanded clay was able to heal crack widths of 0.45–0.46 mm. This suggests that the immobilizing material was able to maintain the bacteria viability and act as a protective material without negatively influencing the healing mechanism. An alternative work [10] reported that using melamine-based microcapsule as an immobilizing material was able to heal a maximum crack width of 0.97 mm.

Several scholars adopted nanosized particles to aid the ability to heal wider crack widths of up to 1.10 mm and 1.20 mm [35,36]. Such promising results might be explained by the ability of nano/microparticles to be uniformly distributable in the concrete matrix, as their significantly small size allows the material to act as a filler material [14,36]. However, it was warned that if the crack width was too large, the healing agent was able to escape into the external environment and as such no healing of the crack would take place regardless of carrier type [15,100]. This is justifiable by the crack surfaces being too far apart to create a bridging bond between them. It is worth noting that as crack width size is an important parameter, natural fibres have been adopted [97] to promote the complete healing of crack widths beyond the thresholds discussed above. Therefore, further research on ways of minimising crack widths utilising immobilization materials may be beneficial and help achieve a more effective self-healing process.

6. Conclusions

Various parameters have been discussed affecting the self-healing mechanisms in cementitious materials with an emphasis on different ways that autogenous and bacteria healing mechanisms can be promoted. The understanding of those parameters could drive the development of new-generation self-healing cementitious materials. To better develop advanced self-healing concretes, the following urgent issues should be reasonably considered.

- Both autogenous and bacteria healing mechanisms cannot be triggered without the presence of water; making water a crucial factor for self-healing to be initiated. This requires the development of healing that doesn't depend on the presence of water as this cannot be always guaranteed in quantities required for healing to be efficient, and the crack will remain present in the material. Therefore, the challenge of developing a way in which moisture in the air can trigger the mechanism may be more favourable trigger.
- Vascular and capsule-based healing are both promising approaches for self-healing cementitious materials. Vascular systems allow for the strategic placement of healing agents within crack zones, as well as providing a wide area in which the healing agent is accessible to heal the cracks. Capsule healing also provides overall healing due to size and dosage of the cracks as microcapsules allow for uniform distribution and macrocapsule show promising results with larger capsules with higher loading capacity which could possibly lead to repeat healing and higher probability of rupture once cracks. Overall, both approaches have their advantages and limitations, and efficacy depending on design, type of healing agent, dosage, crack size and location of cracks require to be considered to produce effective vascular and capsule systems for self-healing.
- The review found that self-healing in cementitious materials using bacteria can be improved by controlling pre-cracking age of specimens, water supply, controlled crack width, bacteria concentration, and immobilization. However, the lack of standardized methods and measuring self-healing have led to variations in experimental results.

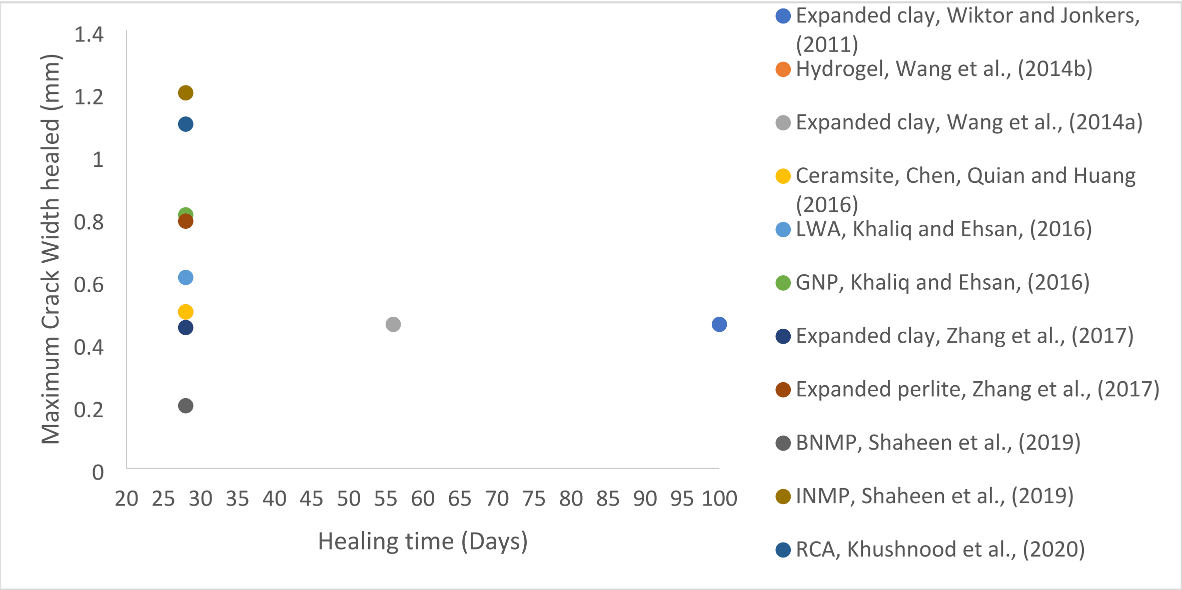


Fig. 12. Graph illustrating maximum crack width healed with time by various researchers using different carrier materials for bacteria-based healing.

Therefore, it is important to establish a standardized method and a way of measuring self-healing to ensure reliable and comparable results across studies conducted in the field.

- Many scholars have found that the direct application of bacteria presents challenges associated with preventing the bacteria from triggering the healing mechanisms when concrete mixing water is added to the mixture, and as viability presented a challenge an option researchers have opted to immobilize the bacteria in porous carrier materials to protect the bacteria.
- To advance self-healing cementitious materials, it is important for attention to shift towards environmentally friendly and economically viable carrier materials and look towards using waste materials that provide multiple services other than just being a carrier. The selection of carrier materials which are energy intensive and has a high cost, which would lead to the price of concrete increasing and add to the current climate emergency.
- A method in preventing the activation of bacteria may be beneficial for the self-healing industry or slowing down bacteria activity during the immobilization of the bacteria and the precipitate medium. To ensure that the bacteria are able to only become activated and germinate once cracks are present. Therefore, the use of colder temperatures can be explored to dry the carrier material rather than using oven drying techniques which could potentially activate the spores.
- It is important to stimulate real-world environments in which the cementitious material will be used in, rather than solely focusing on ideal conditions for healing mechanisms, in which could lead to different performance in material once developed for large scale real element testing. For instance, environments such as ground conditions such as soil can be studied to consider the influence external microorganism can have on the healing mechanism. Therefore, by conducting tests under realistic conditions will allow for the determination of the self-healing systems effectiveness and improve possible limitation.

CRediT authorship contribution statement

Abdulahi Mohamed: Data curation, Conceptualization, Formal analysis, Investigation, Methodology, Writing – original draft. **Yonghui Zhou:** Conceptualization, Methodology. **Elisa Bertolesi:** Supervision, Writing – review & editing. **Mengmei Liu:** Conceptualization, Methodology, Validation. **Feiyu Liao:** Conceptualization, Writing – review & editing. **Mizi Fan:** Conceptualization, Project administration, Supervision, Funding acquisition, Validation, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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