FULL CRITICAL REVIEW

A review of the efficiency of self-healing concrete technologies for durable and sustainable concrete under realistic conditions

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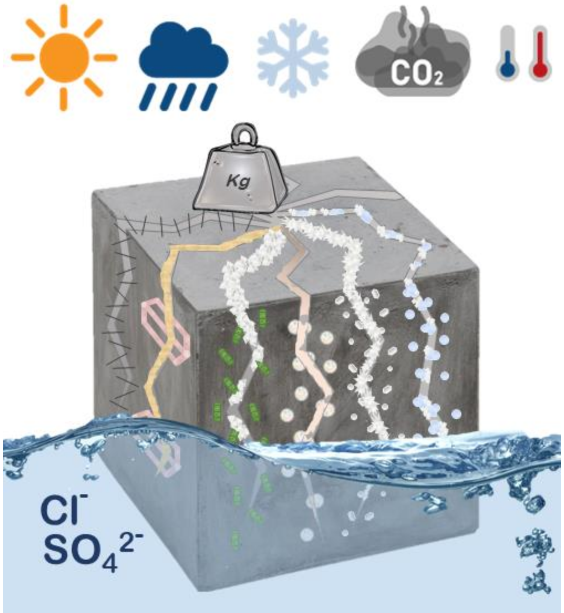
ABSTRACT

Self-healing is recognized as a promising technique for increasing the durability of concrete structures by healing cracks, thereby reducing the need for maintenance activities over the service life and decreasing the environmental impact. Various self-healing technologies have been applied to a wide range of cementitious materials, and the performance has generally been assessed under 'ideal' laboratory conditions. Performance tests under ideal conditions, tailored to the self-healing mechanism, can demonstrate the self-healing potential. However, there is an urgent need to prove the robustness and reliability of self-healing under realistic simulated conditions and in real applications before entering the market. This review focuses on the influence of cracks on degradation phenomena in reinforced concrete structures, the efficiency of different healing agents in various realistic (aggressive) scenarios, test methods for evaluating self-healing efficiency, and provides a pathway for integrating self-healing performance into a life-cycle encompassing durability-based design.



Key-words: Self-healing concrete; crack; durability; sustainability; aggressive environment; degradation; durability indicators; service life.

1. Introduction

Concrete, the most widely used construction material, is intrinsically weak in tension and is quasi-brittle. In many cases, this results in the need to couple it with reinforcement, generally steel. When Reinforced Concrete (RC) structures are exposed to service conditions, including mechanical and environmental actions, tensile stresses may result in the formation of cracks. Cracks impair the functionality/serviceability and durability of RC structures by lowering their stiffness and creating a preferential pathway for the penetration of harmful agents, which will cause degradation of the concrete matrix and the reinforcement. Cracking of concrete can hardly be avoided (unless pre-stressing is applied), but through correct mix proportioning and structural design, in combination with adequate construction and quality control, effective control of crack width and spacing can be achieved. It is thus of utmost importance to study the cracking phenomenon, which will govern the kinetics of material deterioration and the decay of structural performance. This knowledge is essential in planning the frequency and nature of the required maintenance activities. Especially for structures in extremely aggressive environments, rapid and durable repair is needed, the execution of which should consider the accessibility of the degraded area, together with the total repair costs [1].

In this regard, technologies to heal cracks autonomously and hence increase the durability of concrete structures and their service life have gained increasing attention in recent years (Figure 1). The "functionalization" of cementitious materials with self-healing properties was first proposed in the 1990s by Dry [2–4] it was only in 2001 with the article published by White et al. [5] in Nature about self-healing in polymer-based materials, that the interest of both the academic research community and public funding authorities started being steered towards the topic. Four years after this publication in Nature, the Dutch government funded a research program on self-healing materials [6]. Then, other projects with the main focus on self-healing in cementitious materials emerged, in which leading universities in Europe were and still are involved, including SIM-program SHE [7], HEALCON [8], Materials for Life [9], CAPDESIGN [10], LORCENIS [11], SARCOS [12], Resilient Materials for Life [13], EndurCrete [14], ReSHEALience [15] and SMARTINCS [16].

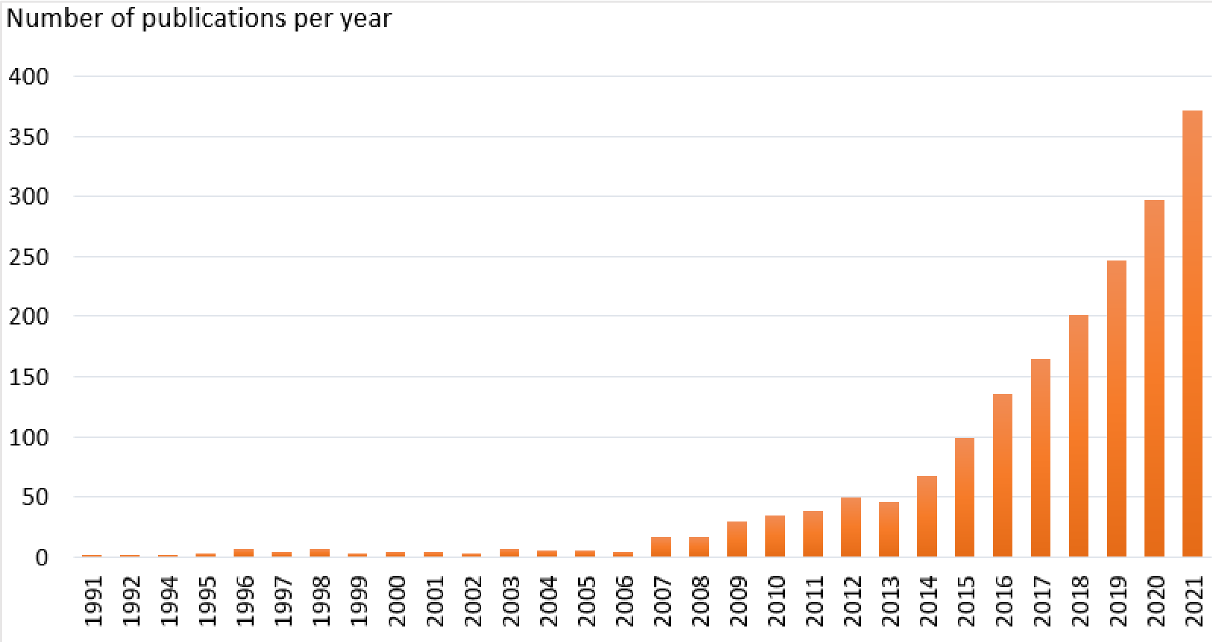


Figure 1. The increasing number of publications on 'self-healing concrete' or 'self-healing' in combination with 'cement' or 'paste' or 'mortar' or 'concrete' or 'cementitious' (Web of Science, 13 April 2022) over the last decades showing that the development and performance assessment of self-healing concrete is a topical subject within the research community.

When discussing self-healing, a distinction has to be made between autogenous healing, which relies on the conventional constituents of the cementitious matrix, and autonomous or autonomic healing, which uses unconventional engineered additions to provide the self-healing function [17]. The most important mechanisms that contribute to autogenous healing when a crack is formed and exposed to water are the chemical processes of continued hydration of yet un-hydrated binder particles near the crack walls, and precipitation of calcium carbonate crystals on the crack faces as a result of the reactions between the calcium ions Ca^{2+} in the concrete matrix and the carbonate ions CO_3^{2-} in the water or carbonic acid formed from carbon dioxide CO_2 in the air entering the crack. Autogenous healing requires the presence of water and is more effective when crack widths are restricted. Therefore, methods to limit crack width (fibre addition), provide water (e.g. addition of superabsorbent polymers), or enhance hydration or crystallization (mineral additions or crystalline admixtures) will be called stimulated or improved autogenous healing.

Autonomous healing includes the application of encapsulated minerals and polymeric healing agents, as well as (encapsulated or granulated) micro-organisms that induce precipitation of $CaCO_3$ as a result of their metabolism and the nucleation properties and zeta-potential of the cell wall ("bacteria-based self-healing concrete"). For encapsulation, microencapsulation, macroencapsulation, and vascular systems have been applied, and this will dictate the size of the damage that can be healed, the repeatability of healing, and the recovery rate [17]. In these systems, the trigger for self-healing is usually the formation of the crack itself, by which the capsules are ruptured and release their content.

Over the past years, several state-of-the-art papers have been published, focusing on the different autogenous and autonomous technologies and their effectiveness [17], the test methods to assess them [18], the mix design of self-healing concrete [19] and the models to predict, also from a design perspective, the outcomes of the phenomenon, including data-driven models derived through Artificial Intelligence (AI) algorithms [17,18,20,21]. Literature surveys provide ample confirmation of autonomous healing of cracks triggered by the crack formation or external stimuli. However, it has to be recognized that much of this evidence was acquired in ideal conditions, often ambient laboratory environments and that, despite the continued efforts, there is still a lack of knowledge regarding the behaviour of self-healing cementitious materials in real structural service scenarios, including extreme conditions or non-steady and non-stress free cracks. This knowledge is of paramount importance for assessing the durability and the robustness of the performance of this new technology [22].

The definition of an ideal condition can vary depending on the healing agent, as exemplified below.

- Crystalline admixtures (CA) show optimal performance under continuous water immersion or, though to a somewhat lesser extent, under wet/dry cycles [23–28].

- Encapsulated polyurethane has the ability to polymerize rapidly in the presence of humidity [29–31]. On the other hand, in unsaturated specimens, the release of a healing agent can be encouraged with the aid of the capillary action by the crack. Temperature changes lead to a variation in polymer viscosity, thus affecting the release process and the effectiveness of crack filling [32].

- Bacteria need water for their metabolic activities and to provoke calcium carbonate precipitation. Aerobic bacteria additionally need oxygen, and spores may not germinate in the deeper part of cracks where available oxygen is limited. Chemical and enzymatic reactions proceed faster at a higher temperature, up to a certain limit [33].

From these examples, it is clear that, in order to make those self-healing technologies reliable and robust for implementation in real structures, efforts to prove the functionality even in adverse conditions, are required [34]. Furthermore, it is important to know how to select the best healing agent as a function of the intended structural service scenario and environmental conditions. Due to the variety of effects that environmental conditions can have on the performance of each healing agent, care must be taken when comparing results from different studies, since degrees of healing can show fluctuations according to environmental conditions [30]. Some research studies considered in-situ applications in order to show that the self-healing techniques developed at laboratory scale could be extended to real situations [3,18,34–40]. However, further research should focus on the long-term durability of the healed structures [17,41], considering more realistic environmental conditions [22,30,31,42–44] including phenomena such as freeze and thaw cycles, salt crystallization pressures, corrosion of reinforcement, long- and short-term actions including cyclic and dynamic ones, which can call for repeated activation of the healing functionality at the same or different crack locations. Moreover, assessing the durability of materials after damage and consequent healing is essential to validate full-scale structural use [31], in a framework in which large-scale self-healing concrete applications are rare and generally show insufficient [45,46] or even unproven efficiency [35,36,47].

Since the use of self-healing technologies in reinforced concrete structures results in longer maintenance-free service lifespans, they have to be evaluated from a life-cycle perspective which encompasses not only technical but also environmental, economic and social aspects [42,48]. In this regard, Life Cycle Assessment (LCA) can help to better highlight the advantages of these cement-based materials with advanced functionalities, evaluating all the possible inputs, outputs and potential environmental benefits over the complete life cycle. Previous studies regarding sustainable concrete solutions have mainly addressed "cradle-to-grave" analysis [49–51]. Only a few have analysed self-healing cementitious composites, sometimes using a "cradle-to-gate" approach, taking into account all the impacts up to the production stage [52–54]. This is due to the lack of data regarding the behaviour of these materials when exposed to real service scenarios and to the lack of environmental inventory information regarding the self-healing agents. Therefore, a wide cradle-to-grave or a cradle-to-cradle LCA analysis is needed to convince the users (designers, contractors, owners) and penetrate the market.

This article represents one of the milestone works of the ITN-MSCA project SMARTINCS funded by the European Commission, with as main objective to review the state-of-the-art on the durability and sustainability of self-healing cementitious materials. This review is conducted with the following purposes: (1) elucidate how cracking influences the transport of aggressive agents and hence the durability of structures in the main aggressive scenarios; (2) review how self-healing mechanisms can increase the durability for each aggressive scenario; (3) give a short overview of methods to evaluate self-healing efficiency; (4) suggest a pathway for incorporating self-healing performance into a life-cycle spanning durability-based design.

2. "Structural durability": influence of the cracked state on degradation phenomena in reinforced concrete structures

The crack formation has to be considered as normal for any reinforced concrete structure under service loads (e.g. Eurocode 2 [55] and fib Model Code 2020 [56]). However, it has to be controlled and limited to an extent which does not compromise the serviceability and durability of the structure. As such, a description of the crack "geometrical" parameters which affect the kinetics of the ingress of aggressive substances and which can be entered into the modelling of material and structural durability is needed. These parameters include crack density, orientation, tortuosity, width, and effective depth, which, together with their self-healing characteristics [57], may all affect the durability of the structure. In international design codes, widely accepted methods are proposed for the calculation of crack width and spacing in ordinary reinforced concrete structures under static loads or restrained deformations. Similar methodologies scarcely exist or are still under development in the case of cracks due to environmental degradation or very early age cracks (e.g. due to plastic shrinkage) or when advanced cement-based materials, such as fibre reinforced cementitious composites, are concerned. Most of all, validation through assessment in real structures for the abovementioned cases is lacking.

The main parameters employed to characterize and design the cracked state of a reinforced concrete element are the crack spacing and the crack width. Structural design codes, including fib Model Code 2020, Eurocode 2 and ACI 318, propose formulations to calculate the crack widths as a function of the reinforcement bar diameter and reinforcement area ratio, concrete cover, concrete tensile strength, steel-concrete bond strength and the stress in the reinforcement induced by the relevant combination of actions. Code formulations allow to predict the maximum width of the crack, e.g. at the intrados for a beam element in flexure; however, it is important to consider that the crack width in such elements varies along its depth. It is apparent that the inherent 3D features of the crack are relevant concerning self-healing effectiveness (the volume of the crack has to be filled and not only the part of the crack close to the crack mouth). Moreover, besides the permeation of aggressive agents in the direction of the crack, which is mainly affected by the maximum crack width, penetration in the direction orthogonal to the crack walls is also of importance, the latter being affected by the evolution of the width of the crack along its depth and by crack tortuosity.

In addition, during the service life of a structure, cracks are affected by the loading and environmental exposure scenario, which leads to evolving characteristics, mainly in terms of crack opening. In fact, structural elements in real-life service conditions experience sustained loads or cyclic actions, which result in through-crack stress states, which may cause the cracks to be "active", meaning unsteady along time. This is of utmost importance also with reference to the long-term performance of healing materials, since the majority of the studies available so far have investigated the performance of healing in specimens that are not subjected to any kind of load and hence under intrinsically static crack conditions. To obtain a reliable prediction of the service life, the complex interaction between physical, chemical and mechanical effects on concrete structures has to be considered. Moreover, different degradation mechanisms can influence each other, and combined effects can be more harmful [58,59].

Table 1 presents a scheme of the main aggressive scenarios with related degradation processes, affected properties of the reinforced concrete and the related indicators. It has been compiled considering the main exposure classes, as reported in EN 206 [60], which discern among: no risk of corrosion or attack, carbonation-induced corrosion, chloride-induced corrosion, freeze-thaw attack, and aggressive chemical environments. For each exposure class, prescriptions are provided related to maximum w/c ratio, minimum strength class, minimum cement content, minimum air content (for freeze/thaw attack) and other specific requirements (e.g. related to types of cement to be used). Specific prescriptions for chemical attack from natural soils and groundwater are based on limiting values for indicators related to the aggressive medium, including pH, contents of carbon dioxide, sulphates, ammonium, magnesium, and soil acidity.

It can be observed that codes currently provide a rather prescriptive approach to durability, referring to the crack width as a durability parameter only in a few exposure conditions. This is because the effects of cracks and crack width on the kinetics of the degradation mechanisms and phenomena are not available in the literature for each exposure scenario. For example, the effects of cracks in structures exposed to aggressive chemical environments (excluding carbonation, chlorides and marine environments) have barely been studied. Degradation by sulphates and chlorides, or combined attack as occurring in a marine environment, has received more attention.

For the whole set of environmental exposure classes, the concrete strength, also related to the w/c ratio [61], which contributes to porosity, resistivity, diffusivity, among other factors, is assumed as a universal concrete performance indicator and hence also as an indicator of durability, at least in its un-cracked state (e.g. high strength concrete is likely to provide higher durability, as result of its lower porosity). Furthermore, the initial calcium hydroxide content is an indirect durability indicator in cementitious materials. It is related to the availability of leachable material and the buffering capacity against carbonation. In addition, the cement content, concrete cover, and chloride content are widely used indicators [62]. Diffusivity, permeability and porosity are extremely important to indicate the durability of concrete structures as they are correlated to transport and diffusion phenomena.

In general, water and gas can carry harmful substances that cause concrete deterioration and/or rebar corrosion. Those fluids have three main transport mechanisms: diffusion, permeation and sorption, but the main ones are diffusion and permeation. Among these mechanisms, permeation is the one that is usually associated with the durability performance of reinforced concrete [63], defining the ease with which a fluid flows through a porous material under a pressure gradient [63]. Permeability is influenced by the size, distribution and continuity of the pores [64], and it is accelerated by the presence of cracks which are considered to be a pathway to aggressive agents. Nevertheless, when harmful substances are present as salts or CO_2, the diffusion mechanism plays the primary role in view of durability. The salt concentration at the surface and the diffusion coefficient of the concrete define the rate of ingress. The diffusion coefficient is highly dependent on the pore structure, mainly affected by the w/c ratio, cement and pozzolan content, curing conditions and presence of cracks. In this respect, the benefits of self-healing concrete are related to their ability to restore the values of the aforementioned indicators to those typical for un-cracked concrete [42].

Investigations regarding self-healing in many of the aforesaid scenarios can hardly be found in the literature. Because of the lack of information, the current review will not discuss acid attack. Nevertheless, it is included in Table 1 as it should be considered as one of the main aggressive scenarios. The following sections provide a comprehensive review regarding the current knowledge on the effects of cracks on the degradation mechanisms and the behaviour of self-healing concrete in those scenarios.

Table 1. Degradation process and durability indicators for each aggressive scenario.

<table>
<tr><th>Main Aggressive scenarios</th><th>Type of attack</th><th>Degradation process in RC</th><th>Effect on RC</th><th>Specific durability indicators (even in SH concrete)</th><th>General durability indicators</th><th>Concrete quality indicators</th></tr>
<tr><td>Chloride attack</td><td>Chemical attack</td><td>Salt precipitation
Corrosion
Pitting
Cracking</td><td>↑Porosity
↑Diffusivity
↑Potential gradient
↓Resistivity
↑Acidification ↓pH</td><td>Chloride Penetration
Corrosion Potential</td><td rowspan="8">Water accessible porosity

Diffusion coefficient

Gas/water permeability

Portlandite content

Capillary absorption

Electrical resistivity</td><td rowspan="9">Strength

w/c ratio

Cement content

Cover depth

Chloride content

Porosity</td></tr>
<tr><td>Carbonation</td><td>Chemical attack</td><td>Rebar depassivation
Corrosion
Cracking</td><td>↕Porosity
↕Permeability
↕Diffusivity
↑Potential gradient
↓Resistivity
↑Acidification ↓pH</td><td>Carbonation rate
Corrosion Potential</td></tr>
<tr><td>Sulphate attack</td><td>Chemical/ Physical attack</td><td>Expansion
Internal tension
Mechanical softening
Cracking
Mass loss
Flaking
Scaling</td><td>↑Porosity
↑Permeability
↑Diffusivity
↑Internal damage
↑Potential gradient
↓Resistivity
↑Acidification ↓pH</td><td>Volume changes
Mass loss
Sulphate content</td></tr>
<tr><td>Combined attack (marine environment)</td><td>Chemical/ Physical attack</td><td>Expansion
Internal tension
Mass loss
Cracking
Corrosion</td><td>↑Porosity
↑Permeability
↑Diffusivity
↑Internal damage
↑Potential gradient
↓Resistivity
↑Acidification ↓pH</td><td>Chloride Penetration
Corrosion Potential
Volume changes
Mass loss</td></tr>
<tr><td>Acid attack</td><td>Chemical/ Physical attack</td><td>Leaching of hydrated products
Cracking</td><td>↑Porosity
↑Permeability
↑Diffusivity
↑Internal damage
↑Potential gradient
↓Resistivity
↑Acidification ↓pH</td><td>Portlandite content
Mass loss
Volume change</td></tr>
<tr><td>Freeze-thaw attack with de-icing salts</td><td>Chemical/ Physical attack</td><td>Scaling
Internal tension
Cracking
Corrosion</td><td>↑Porosity
↑Permeability
↑Diffusivity
↑Internal damage
↑Potential gradient
↓Resistivity
↑Acidification ↓pH</td><td>Mass loss
Chloride Penetration
Corrosion Potential</td></tr>
<tr><td>Freeze-thaw attack without de-icing salts</td><td>Physical attack</td><td>Internal tension
Cracking</td><td>↑Porosity
↑Permeability
↑Internal damage</td><td>Ultrasonic velocity
Acoustic emission
Modulus of elasticity</td></tr>
<tr><td>Water pressure</td><td>Physical attack</td><td>Leaching of hydration products</td><td>↑Porosity
↑Permeability
↑Internal damage</td><td>Water permeability under pressure</td></tr>
<tr><td>High/low temperature and temperature variation</td><td>Physical attack</td><td>Shrinkage
Cracking</td><td>↑Permeability
↑Internal damage
↓Resistivity</td><td colspan="2"></td></tr>
</table>

Note: SH: Self-healing, RC: reinforced concrete, the arrows mean: ↑ increase, ↓ decrease and ↕ increase or decrease.

2.1 Chloride attack

Marine structures, bridge decks, road slabs and parking structures are the most common concrete constructions being susceptible to corrosion induced by chlorides. Chloride ions originate from various environmental sources to which the structure is exposed during its service life, including seawater, marine airborne salts and de-icing salts. A thermodynamic service life model was proposed by Tuutti in the 80´s [65] for reinforcement corrosion that highlights two phases: initiation and propagation. In the initiation phase, two sub-periods have been derived for the determination of the time to chloride-induced corrosion: (1) the time for the transport of chloride ions penetrating from the environment through the concrete cover to reach the rebar level, and (2) the time for accumulation of chloride at the rebar level up to a critical content (chloride threshold) which allows active corrosion to start. From then onwards, pitting corrosion propagates. This model concept has been quantified and incorporated in the standards for the service life calculation of concrete structures, such as *fib* Bulletin 34 (2006) [66], *fib* Bulletin 53 (2009) [67], Eurocode 2 (2011) [55], EHE-08 (2008) [68], ENV 206 (2016) [60], Spanish

Structural Code (2021) [69]. Standards refer to a chloride threshold expressed as % of total chloride by weight of the binder, assuming that bound chlorides could be released, as can be the case upon carbonation. In reality, the threshold value of chloride for corrosion initiation has been found to show a large scatter [70,71]. It depends on several factors characterizing the steel-concrete interface, as those mentioned in Angst et al. [72,73]. Construction codes generally consider the limit of 0.4% total chloride by cement weight. In contrast, in the recent Spanish structural code (2021) [69], different values are included as chloride thresholds depending on environmental exposure. Moreover, the rate of chloride ingress is affected by the quality of the concrete cover, chloride concentration and exposure conditions [74–76].

The presence of cracks in the concrete cover provides easier access for the chloride ions to the deeper parts of the concrete [77,78] (Figure 2), causing corrosion to initiate in a shorter period [77]. The chloride ingress will increase with the increase of micro-crack density independently of the concrete characteristics [79]. Moreover, the chloride attack could be aggravated by salt crystallization pressures in the crack [80]. In chloride-rich environments, the crack width limit should be more restrictive than for environments free of chlorides since chloride ions have a small size and high diffusion capacity [81,82]. The crack geometry, including width, depth and tortuosity, affects the transport of chloride ions [83–85]. The wider and deeper the crack, the earlier the reinforcement corrosion will occur [83]. Maes [86] stated that 10 µm is the critical crack width, above which chlorides penetrate more rapidly through the crack than in the un-cracked zone, whereas Van den Heede et al. [87] suggested that for a crack width lower than 50 µm, chloride penetration through the crack would be negligible. Russo et al. [85] found that chloride penetration increased at least 1.5 times for concrete with different characteristics when increasing the crack width from 5 to 70 µm and crack depth to 40 mm. Djerbi et al. [88] found that the chloride diffusion coefficient for crack widths less than 80 µm was higher for conventional concrete than for high-performance concrete (HPC) due to its higher porosity. However, for crack widths larger than 80 µm, the diffusion coefficient did not depend on the crack width or material characteristics. Kušter Marić et al. [89] measured the chloride content at different locations of a concrete bridge in service. They presented 100 µm as a crack width limit beyond which depassivation time was significantly reduced, independent of the cover thickness.

Francois and Maso [77] noticed a higher chloride diffusion in the tensile zone than in the compression zone of beams after flexural loading due to the damage at the grain/paste interface. This was confirmed by Gowripalan et al. [90], who related the difference to cracks in the tension zone and a decrease of porosity in the compression zone [90]. There is an evident lack of information regarding the effect of cracks on the chloride threshold for reinforcement corrosion onset. Pettersson et al. [91] found that for a high crack width/cover ratio, the chloride threshold decreased in the seawater splash zone for high-strength concrete containing silica fume.

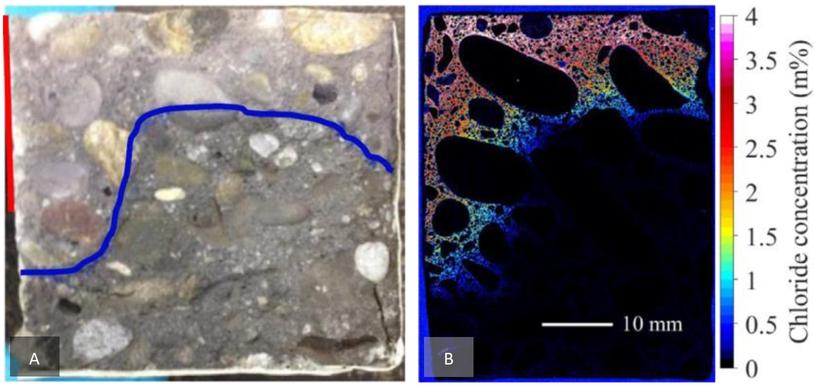


Figure 2. Cracks in concrete provide easy access for chloride ions. The red line indicates the crack location. Chloride ingress can be visualized by spraying with 0.1 M $AgNO_3$ resulting in white AgCl precipitation - the blue line denotes the discolouration front (A). The chloride element map shows the concentration gradients by electron probe micro analysis (EPMA) (B). [78] *Copyright 2019, Elsevier.*

2.2 Carbonation

The alkaline nature of the concrete facilitates its interaction with environmental CO_2 decreasing the pH of the cover. When carbon dioxide from the air comes in contact with the concrete, first, it has to be dissolved in the pore water to form HCO_3^- ions. The dissolved CO_2 reacts with portlandite (CH) and C-S-H gel to produce calcium carbonate and decrease the pH of the pore solution. That results in depassivation of the reinforcement and enables rebar corrosion if enough moisture is available in the environment. The rate at which this process occurs depends mainly on concrete and environmental characteristics [92,93]. A recently published literature review by RILEM TC 281-CCC presents the effect of concrete characteristics, relative humidity, temperature, CO_2 concentration, age and curing conditions on the carbonation mechanism, kinetics and microstructural alterations in cementitious systems, including those containing supplementary cementitious materials (SCM). Moreover, the effect of carbonation on the transport properties and porosity of concrete with SCMs was studied [94].

Cracks can favour the CO_2 penetration through the concrete cover, thus triggering an earlier start of the reinforcement corrosion. Alahmad et al. [95] studied for mortars the effect of cracks with openings ranging from 9 to 400 µm on the ability of CO_2 to diffuse along the crack. After accelerated carbonation (50% CO_2) for 65 days, at 23°C and RH 65 %, they found that crack opening greatly impacted the carbonation. For crack widths higher than 60 µm, the carbonation depth perpendicular to the crack was similar to carbonation at an outer surface, indicating that CO_2 penetrates freely in the crack. However, for cracks below 41 µm, the carbonation perpendicular to the crack progressively decreased. For crack widths of 9 µm, carbon dioxide diffusion through the crack was stopped, which was attributed to self-healing by carbonation products. Van Mullem et al. [96] showed that carbonation along realistic cracks in mortars was significantly affected by the crack tortuosity once crack widths went below 100 µm. Han et al. [97] claimed similar results in concrete after accelerated carbonation at 20% CO_2, at 20 ± 2 °C and RH 70 ± 5 %. For crack widths bigger than 100 µm, the carbonation depth perpendicular to the crack walls was similar as for the concrete surface (Figure 3). Bogas et al. [98] found for structural lightweight aggregate concrete (LWAC) that the crack opening (100-300 µm) was less relevant in comparison with regular weight concrete since porous lightweight aggregates anyhow led to an increase in the carbonation rate. Carević and Ignjatović [99] claimed that the presence of small cracks (50 µm) in reinforced concrete leads to a twice higher carbonation depth after exposing to a CO_2 concentration of 2% for 28 days. The average carbonation depth increased when the crack width increased.

There is an evident lack of knowledge regarding corrosion initiation risk due to the crack presence and the carbonation effect. Marcos-Meson et al. [82] exposed steel fibre reinforced concrete (SFRC) specimens with 50 to 300 µm cracks to wet-dry cycles involving both chloride and carbon dioxide for one and two years. Steel fibres directly exposed at the crack were corroded since the pH at the crack walls decreased to about 9 due to carbonation. However, the impact on the corrosion of reinforcement was limited due to the precipitation of calcite and hydration products partially sealing the cracks, thus slowing down the further carbonation process.

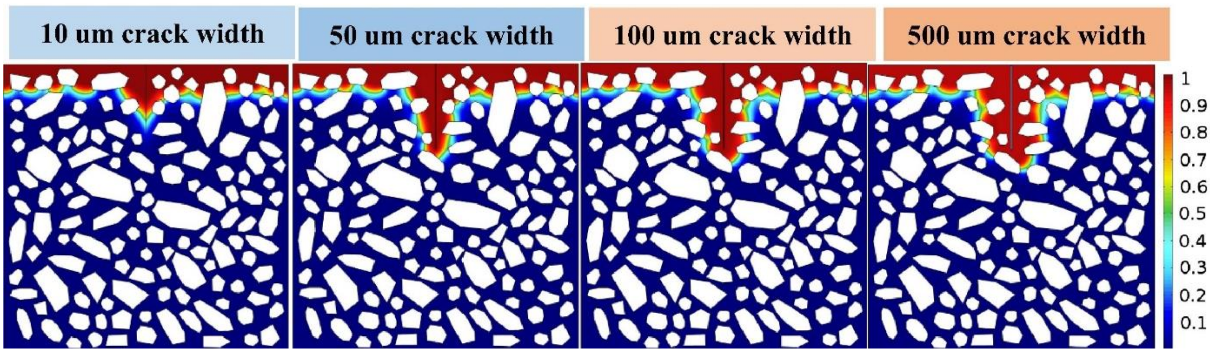


Figure 3. Relative carbonation content after 10 years in cracks with different crack widths using the virtual COMSOL Multiphysics model proposed by Saetta [100]. [97] *Copyright 2016, Elsevier.*

2.3 Sulphate attack

Each salt induces a different level of deterioration related to the solubility of its reaction products, the reaction kinetics and the nature of the crystallization pressure of solid products, with sulphate being the most harmful to concrete [101]. Examples of structures which can be subjected to sulphate attack include basement walls, piers, abutments, and tunnels, which are in contact with ground water, soil, seawater, industrial effluents, sewage water or waste water [102,103]. Depending on the sulphate exposure condition, the attack is categorized as a chemical or physical phenomenon [104,105]. Total immersion ends in chemical attack, while physical attack prevails when surfaces are (temporarily) exposed to drying conditions [106]. The physical salt attack is connected with continuous surface scaling and flaking of concrete due to salt crystallization in the pores adjacent to the drying surface [107].

On the other hand, a chemical salt attack results in a direct reaction with the hydrated cement paste that leads to mechanical softening, expansion, cracking, spalling and disintegration [108,109]. Overall, these processes are related to the entry mechanism that can be diffusion or permeation when water contaminated with sulphates penetrates the concrete. Nehdi et al. [104] commented that studying only chemical attacks, as is generally done in experimental research, does not represent the situation in most concrete structures with some area exposed to the air.

Sulphates are mostly found as sodium sulphate (Na_2SO_4) or magnesium sulphate ($MgSO_4$) but can also have calcium (Ca) or iron (Fe) as the cation, leading to a different attack mechanism and kinetics [110]. For Na_2SO_4, secondary gypsum and ettringite precipitate with a volume expansion which can lead to an intense cracking process because of internal stresses [111], leading to increased permeation of the sulphate solution. While tricalcium aluminate (C_3A) and calcium hydroxide (CH) are attacked, the calcium silicate hydrate (C-S-H) is kept intact. However, considering $MgSO_4$ attack, thaumasite and brucite can be formed, causing more impact on the mechanical resistance [110]. At first, the brucite layer slows down the sulphate attack, which slowly continues by diffusion across the surface layer [111]. As the attack progresses, expansion results in cracking of the brucite layer, further decalcification of the C-S-H and ultimately conversion to the non-cementitious magnesium silicate hydrate (M-S-H) [111].

Idiart et al. [112] progressed in a model prediction considering the presence of cracks and further degradation by sulphate attack. They included diffusion through the cracked concrete based on the study by Djerbi et al. [88], defining a linear relation between the crack width and the diffusion coefficient in cracks up to 100 µm. Cracks larger than 100 µm had diffusion similar to diffusivity in free solution. Liu et al. [113] analysed fatigue loading, showing that the created cracks accelerate the process of deterioration by sulphates.

Overall, the major factors influencing the sulphate attack are the available amount of sulphate ions, permeability of concrete, quantity of C_3A, type of binder, pH, temperature and type of cation [86,105,114]. The first stage of the attack involves the formation of expansive products leading to the second stage where cracks appear and where crack formation advances proportional to the rate of expansion reactions until complete deterioration [114,115], which can be worse if combined with a physical attack [104]. The presence of cracks accelerates the sulphate ingress and is the main transfer path in the later stage [105].

2.4 Combined attack (including marine environment)

Academic research tends to focus on isolated effects of a single aggressive agent, and only a few studies have evaluated combined attack actions. Still, the topic has been getting more and more attention over the last years. Most real situations are likely to be characterized by combined attack of different aggressive species involving different interacting degradation mechanisms. For example, Meng et al. [116] considered the combined effect of sulphate attack and freeze-thaw actions. They identified that sodium sulphate accelerates concrete damage under freezing and thawing compared to tap water. Li et al. [117] also reported that concrete deterioration is aggravated when freezing-thawing is combined with sulphate attack. Gong et al. [118] investigated the expansion and mechanical deterioration of concrete damaged by coupled alkali-silica reactions (ASR) and freeze-thaw cycles (FT). They verified that the silica gel formed by ASR could partially restore the damages due to the FT showing a positive effect on the mechanical properties. Liu et al. [119] conducted a numerical analysis for combined load-induced cracking and chloride transport. They identified that longer cracks facilitate the chloride ingress with higher concentrations at the crack tip.

Among all combined attack scenarios, the one with the greatest coverage is the marine environment, where the main salts, sodium chloride and magnesium sulphate, act combined. The deterioration of concrete in the marine environment is related to the saturation degree (submerged, tidal or atmospheric zone), affecting the attack mechanism (chemical/physical) and the transport of the aggressive agents. On the other hand, whereas the seawater changes considerably in salinity in different areas, the type of ions and their proportions are similar [120].

Maes [86] considered combined attack by simulated seawater and investigated the effect of chlorides on sulphate attack as well as the effect of sulphates on chloride attack. In the case of magnesium sulphate attack, a large dependency on the environmental temperature and binder type was observed, while sodium sulphate attack mainly was related to binding with the C_3A hydration products. When considering chloride ingress and the risk of reinforcement corrosion, the influence of sulphates was affected by the exposure time. In the first stage, chloride diffusion was barely affected by the presence of sulphates. In the second stage, the chloride diffusion decreased with the sulphate content since voluminous sulphate-related reaction products obstructed the pore network. In the final stage, chloride diffusion in the presence of sulphates increased since the continued growth of the expansive products leads to crack formation. Zhao et al. [121] confirmed that a combined sulphate-chloride attack aggravates the damage because an increasingly severe cracking process opens paths for chloride ion penetration. Furthermore, chloride binding decreases when sulphate content increases, even worse for higher temperatures when the diffusivity increases [122,123].

Sun et al. [124] examined the steel corrosion and cracking process in cement paste exposed to combined sulphate-chloride attack by microtomography (µCT) analysis. The cracks formed due to the sulphate attack occurred prior to the cracks formed by corrosion; moreover, the chloride ions mitigated the sulphate attack delaying the development of cracks. Wu et al. [125] found that the presence of chlorides can mitigate the effect of magnesium sulphate attack and delay the deterioration in the early stage, but it can promote the migration of sulphates inside the concrete in the later stage. Once the formation of expansive products has resulted in cracks, these trigger more severe chloride-induced corrosion of steel reinforcement [89]. Kušter Marić et al. [89] verified that cracks larger than 100 µm decrease the time for depassivation significantly, despite the thickness of the cover zone. Whereas in un-cracked concrete with a sufficient cover, a service life of 100 years is considered by the European standards, in this study, the cracked concrete in the marine environment had its service life reduced to one year (chloride threshold exceeded at the reinforcement). A large-scale experimental program on concrete with blended cement during 18 years considering submerged, tidal and atmospheric zones [126] showed that the expansive reaction products (brucite, ettringite, calcite) could fill cracks and pores, preventing further chloride ingress at first instance. However, as the attack persisted, the decalcification of C-S-H increased the total porosity resulting in further degradation.

Montes et al. [127] indicate that corrosion caused by marine exposure tests was aggravated by increasing the crack width from 250 µm to 500 µm. Otieno et al. [128] noticed by a long-term experimental program with cracked concrete subjected to a tidal zone that the cover thickness can prevent corrosion in narrow cracks. Nonetheless, for wider crack widths of 400 and 700 µm, the cover thickness did not have a considerable effect. Lopez-Calvo et al. [129] revealed a clear increase of corrosion potential with crack width in a simulated marine tidal condition. However, with a higher cover thickness, the limits on crack widths could be increased: for a cover of 25 mm, corrosion was noticed for cracks wider than 90 µm, whereas for a cover of 45 mm, this was only the case for cracks above 180 µm. Du et al. [130] detected by mimicking a marine exposure that 700 days were needed to cause corrosion in cracks with a width of 100 µm; on the other hand, while this was reduced to 460 days for cracks of 300 µm.

Studies on advanced cement-based materials are more scarce. Lv et al. [131] verified the chloride ion transport in microcracked ultra-high performance concrete (UHPC) with steel fibres in a marine environment. They observed that even cracks around 50 µm wide formed in the protective layer over the steel fibres become rich in chloride ions. Taking that into account, the corrosion products on the steel fibres can produce further stress internally, resulting in tiny cracks in the surrounding matrix.

2.5 Freeze-thaw attack with or without de-icing salts

In freezing conditions, concrete will crack when the microstructure cannot support the stress caused by water expansion, and stress release upon cracking will open additional space for water which will expand again during freezing [132]. There are two types of deterioration due to freezing and thawing (FT): internal cracking, which is the main problem in the absence of chlorides and surface scaling, which is related to the presence of de-icing salts [133]. The internal damage is controlled by the degree of pore saturation, which depends on the bulk moisture uptake [134,135]. Shields et al. [136] varied degrees of saturation under FT cycles and detected higher crack volumes with increased saturation levels. Besides the saturation, Peng et al. [132] identified that the temperature distribution greatly influences FT's effect. Overall, three factors play an essential role, namely material permeability, which is related to the composition and the presence of cracks; the spacing factor between the capillary pores and air voids; and the rate of freezing [133]. Recently, Wang et al. [137] reviewed the three main essential factors to achieve a better performance in FT conditions: (1) improving the internal pore system, (2) reducing the water absorption of the concrete, and (3) controlling the cracking process.

The first, namely the internal pore system (1), is enhanced mainly by the use of an air-entraining agent but also by the use of supplementary cementitious materials and nanoparticles [137], beyond the reduction of the water/cement ratio [138,139]. A proper air void distribution is of significant importance to alleviate the expansion by the freezing water [140]. Şahin et al. [139] claimed that the adequate spacing factor between air voids depends on the freezing condition, i.e., the rate of freezing, the minimum temperature and the duration at the minimum temperature. The second factor mentioned by Wang et al. [137] is the reduction in water absorption (2), which means a lower permeability [133,141], and hence a reduced scaling of the surface layer [142]. Though scaling is superficial and does not compromise mechanical integrity, the weakened surface layer becomes susceptible to further ingress of aggressive agents affecting the durability [143,144]. Crack control (3) is the last factor that can contribute to the FT resistance and which might be improved by using fibres and nanoparticles. Zeng et al. [145] proved that steel fibres increased the FT resistance by bridging cracks and controlling their width, hence decreasing the permeability in the cracked state. Wu et al. [146] applied cellulose/PVA hydrogels to reduce the free water content, avoid crack propagation and hence decrease the effects of the FT.

The presence of de-icing salts on the surface of concrete structures at freezing temperatures causes severe scaling damage, intensified by the further salt solution penetration into the pores [63,93,143,147,148]. Salts change the damage development due to crystallisation and melting temperature variation [149]. Ma et al. [150] illustrated for three types of de-icing salts, ethylene glycol, NaCl and calcium magnesium acetate how spalling started when reaching the critical superficial crack density of 85 to 88%. Figure 4 presents how the cracking behaviour progresses in concrete without initial cracks and with a preliminary cracking process [151]. The presence of cracks accelerates the water absorption, increasing the concrete saturation. Then the freezing and expansion process induces tensile stress. After several FT cycles, the width and quantity of the cracks increase until the worst-case scenario of scaling is reached. Li et al. [152] made a numerical study analysing chloride penetration in cracks induced by freezing and thawing. The authors concluded that the cracks affect the chloride ingress, which is further increased with the number of FT cycles as a result of the crack propagation. Cracks below 50 µm had a small effect, while cracks between 50 µm to 100 µm increased the chloride penetration significantly.

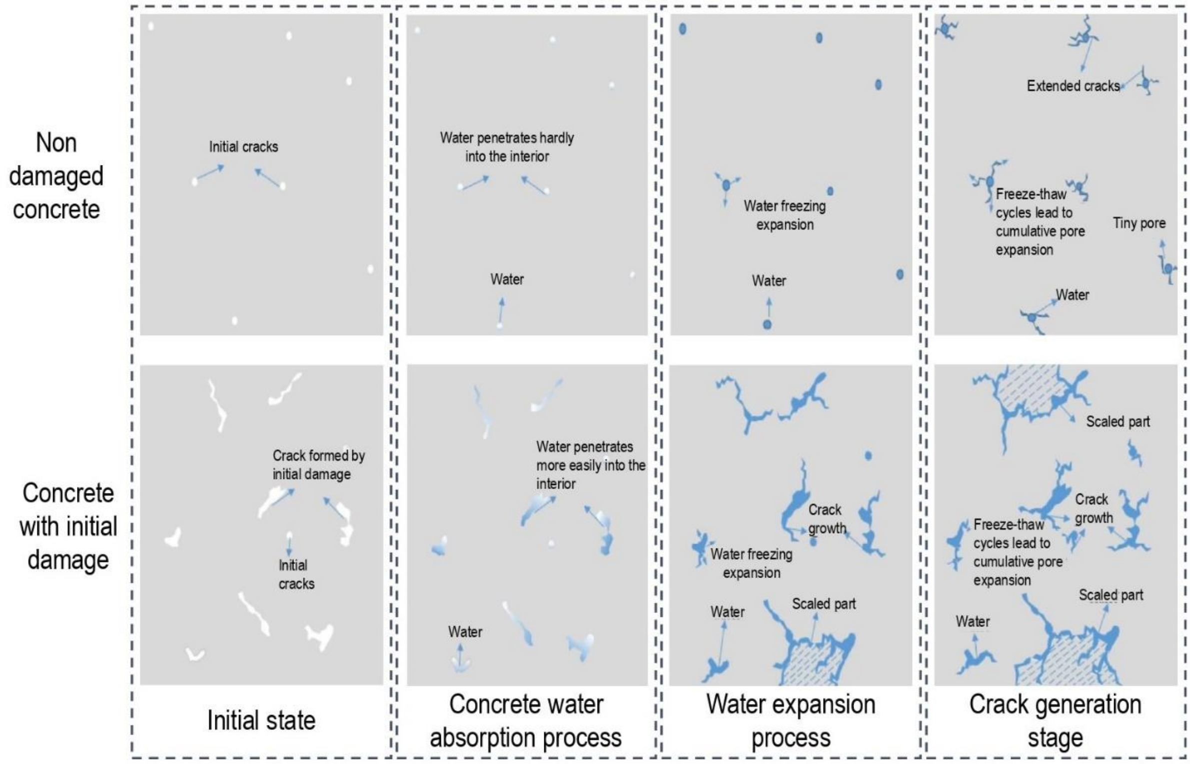


Figure 4. The degradation caused by the internal tension leads to cracking propagation in concrete without damage (top) compared to concrete having prior cracks (bottom) under freezing and thawing cycles. [151] *Copyright 2020, Elsevier.*

2.6 Water pressure

Dams, water tanks, reservoirs, swimming pools, and structures in deep water such as port and harbour installations, undersea tunnels, offshore oil platforms, and caissons are examples of structures that should sustain hydrostatic pressure loads. The hydrostatic pressure causing water pressure inside cracks might reduce the ultimate resistance of the structure significantly [153]. Huge hydraulic loads can accelerate the propagation of cracks in concrete structures and increase the damage caused by hydraulic fracture [154]. With this concern, concrete has to be adopted with a higher capacity to control crack propagation and permeability, even under sustained loadings. It should be emphasized that for immersed structures (deep or shallow), the durability (in normal conditions) is mostly not a significant issue, predominately because of the low O_2 levels. Hence corrosion takes a very long time to initiate and a very long time to propagate. What is of major importance though is the stability of the cracks and the structural integrity, especially in deep water installations. High-performance fibre reinforced cementitious composites (HPFRCCs) can be the ideal candidates in these scenarios, mainly because of their ability to spread otherwise localized damage into narrow and tightly spaced multiple cracks [155].

Moreover, water under pressure can foster the penetration of aggressive agents, and permeability is the most important indicator of durability. For example, in structures in deep seawater, the diffusion mechanism of chlorides becomes less important than penetration under water pressure. In underground structures, including foundations, the water pressure is affected by the surrounding soil and the high water level that may intensify sulphate attack. Xiao et al. [156] considered a mimicked seawall where both sides of the structure were affected by water pressure. In this case, the chloride ingress at the seawater side was prevented because of the water pressure of 0.3 to 0.9 MPa on the other side of the wall. Brzozowsk and Horszczaruk [157] clarified the influence of hydrostatic pressure on the adhesion of repair materials.

Water pressure has an important role in crack propagation. Wang et al. [58] developed a numerical model considering crack propagation in dams under static and seismic loads, showing that the water pressure within the crack contributes to an increasing crack length. Cui et al. [158,159] found that the concrete compressive strength and bulk modulus were reduced with an increase in hydrostatic pressure between 30 and 500 MPa, due to the increased volume of microcracks in the aggregate-mortar interfacial transition zone (ITZ). Yi et al. [160] verified for cracked concrete up to 2 MPa water pressure that the water transport increased with crack width and hydraulic pressure. Cracks of 30 µm and 50 µm had similar flow rate, while a crack width of 100 µm increased the flow rate significantly. Indeed, Edvardsen [161] has shown that the flow rate scales with the third power of the crack width.

2.7 High/low environmental temperatures and temperature variations

The temperature of the environment can vary significantly, leading to the expansion and contraction of structural elements, which can affect their durability. Cold weather during winter times can result in negative temperatures for long periods. However, freezing conditions were already treated in section 2.5 and will not be discussed here. Also, fire damage related to spalling caused by the development of internal steam pressure, dehydration and phase-changes of hydration products will not be treated. Still, structures subject to solar radiation can reach high temperatures on the exposed surface (around 60°C) on hot days. In sandwich panels, the difference between the internal and external temperature can reach more than 20°C [162] as a function of the thermal diffusivity and conductivity of concrete. These depend essentially on the water content in the concrete, the type of aggregate, and the proportion of aggregates in the mixture [163], whereas the effects of the presence of cracks can be considered as negligible [164].

The volume variations resulting from temperature cycles and the temperature gradients inside a structural element may generate tensile stresses in the matrix resulting in cracks, because of the likely presence of external and internal restraints, including reinforcement. Structures must hence be designed with details allowing those movements, such as expansion joints, and employing concrete that can effectively control the consequences of such deformations, including, for instance, fibre reinforcement to control the crack opening [165].

3. Self-healing under aggressive environmental conditions

Most of the laboratory studies dealing with self-healing of concrete do consider 'ideal' conditions regarding the healing regime. And while the definition of the 'ideal' condition is obviously dependent on the healing technology considered, as will be explained further, all 'ideal' conditions have a common denominator: (1) cracks are stress-free upon healing and (2) young specimens are subjected to a single cracking/healing event. This does not match with reality, so it will be extremely important to extend the work from 'ideal' conditions to 'real' conditions and prove the performance of self-healing concrete in real situations in order to allow self-healing to penetrate the market further. Starting from healing under 'ideal' conditions, in the next sections, we will gradually move away from it, considering the stress state through the crack and the stability of the crack over time as well as the stability of the healing functionality over time. In the end, healing under different aggressive conditions is addressed.

3.1 Ideal conditions and water availability

Ideal conditions vary according to the self-healing mechanism considered. For autogenous self-healing or improved autogenous healing by including crystalline admixture (CA) or supplementary cementitious materials (SCM), it is well known that there is a need for water. Either high humidity or preferably wet-dry cycles or immersion in water, are needed for developing the best performance [17,23–26,166,167]. Generally, autogenous healing will be limited to relatively small crack openings [17,18,42]. There is evidence of crack healing between 10 to 100 µm, sometimes up to 200 µm, only in the presence of water [17].

If in real situations, the structure is not in contact with rain or other sources of liquid water, these mechanisms may become less effective or even useless [22]. Some agents, such as superabsorbent polymers (SAP), can contribute to the capture of moisture from the air and hence serve as an activator for autogenous healing [168]. The crack sealing provided by the immediate swelling of SAP particles in contact with moisture, and the activation of autogenous healing by releasing this moisture to unhydrated binder particles near the crack wall, can contribute to the self-healing mechanism. The effect of the humidity level is reflected in the effectiveness of crack closure. As shown by Snoeck et al. [169], in environments with a relative humidity higher than 60%, only samples with SAPs showed visual closure of the cracks, indicating that SAP particles have hygroscopic capacity and can attract moisture from the environment. Samples containing SAPs and exposed to wet/dry cycles could close cracks completely up to about 140 µm, while cracks larger than 200 µm showed only partial healing [169]. To guarantee healing efficiency, it is necessary to keep the crack opening below the threshold of the healing agent. Crack width reduction through fibre reinforcement will enable SAPs to seal off the matrix from penetration of aggressive agents even in environments with less available water [17,169,170].

The behaviour and triggers required for healing of concrete with CA are similar to those required for autogenous healing. Thus, it has been highlighted that the use of CA as a healing agent is limited to structures that have water availability [26,171,172]. Structures such as water tanks and reservoirs, submerged structures or structures with abundant water availability can be healed with CA. Cementitious matrices with CA have a higher pH and release more Ca^{2+}, favouring the precipitation of calcium carbonate and thus the crack closure [23]. Borg et al. [26] reported that under ideal conditions of submersion, cracks between 100 - 150 µm closed in 56 days, cracks of 150 - 300 µm closed in 84 days, while complete closure was not achievable for cracks above 300 µm. Without water contact, CA could not fully close the cracks under 300 µm in 42 days, even with high relative humidity between 95-100% [25,172]. Though, some CA demonstrated the capacity to catalyse the healing reactions even by exploiting the atmospheric humidity [24]: a regular concrete, upon exposure to humid climates (RH > 65%), showed a healing capacity equal to the autogenous healing exhibited by the reference without CA addition when permanently submerged in water.

The use of fibres in engineered cementitious composites (ECC) [173–176] as well as in ultra-high-performance fibre-reinforced concrete (UHPFRC) [177–179] or other cementitious matrices brings great benefits for self-healing. First of all, these materials benefit from their high autogenous healing capacity due to their composition (high binder content, high content of supplementary cementitious materials featuring pozzolanic or late cementitious action, and low w/b ratio). Moreover, the fibres contribute to redistributing the stresses after cracking and are instrumental in generating cracks with a smaller width, so the damage, otherwise localized into a single wider crack, is spread into a larger number of more tightly spaced and narrow cracks. These cracks have a greater potential for closing, which promotes recovery of both durability and mechanical properties, since the ratio of available healing agent to crack volume for smaller cracks is greater than in wide cracks. After ten wet-dry cycles (submersion in water at 20°C for 24 h and drying in laboratory air at 21 ± 1°C, 50 ± 5% RH for 24 h), Li and Yang [173] reported that cracks of up to 50 µm could be fully healed autogenously with complete restoration of mechanical capacity and prevention of the transport of fluids. Guan et al. [176] analysed an ECC with limestone powder, noting increased self-healing because of crack control by the fibres and contribution from the limestone powder by forming nucleation sites accelerating the hydration product formation, and increasing calcium carbonate precipitation. Hung and Su [174] showed that healing allowed ECCs to regain strength and stiffness effectively. Ferrara et al. [177,178] highlighted that for High-Performance Fibre Reinforced Cementitious Composites (HPFRCCs), the presence of water from high air humidity or wet and dry cycles enabled the autogenous healing, even after periods as long as two years.

A bacterial self-healing concrete has requirements which depend on the characteristics of the microorganisms that are used. The aerobic bacteria proposed by Jonkers et al. [180], for example, need in addition to water also the presence of O_2, which can be limited deeper down in the cracks. Alternative methods can be used in case of a lack of O_2, such as using nitrate-reducing bacteria together with nitrate as an alternative electron acceptor that enables bacterial $CaCO_3$ precipitation without O_2 being available [41,48,181]. Protective means for these organisms are necessary to guarantee survival within the hostile environment of a cement matrix. Several technologies for the protection of bacterial cells or spores have been used. Absorption of bacterial spores into expanded clay particles guaranteed the microbiologically induced calcite precipitation (MICP), but resulted in a reduction of mechanical resistance [182,183]. A new granulation technology that helps to protect nitrate-reducing bacteria called ACDC (active compact denitrifying core) revealed that these granules are compatible with concrete and allow the bacteria to survive in mortar. When added as 1% w/w cement, after 28 days of water immersion, they induced complete crack closure up to 500 µm crack width and a 74% reduction of water absorption. The crack filling was noticed in the internal volume of the crack and not just near the crack mouth [41,48]. Mortars with 0.5% ACDC w/w cement showed healing of cracks up to 400 µm for 95%, which led to an 80% decrease in water permeability [184]. A similar method of granules containing ureolytic bacteria called CERUP (Cyclic EnRiched Ureolytic Powder) or MUC (Mixed Ureolytic Culture) achieved closure of cracks with a width of 450 µm after 28 days immersed healing regime [185]. Among the published works, the largest local crack closure from bacterial activity in ideal conditions was 970 µm, about four times that of the non-bacteria control series [186]. Generally, cracks around 350 to 500 µm can be closed by means of bacterial activity under ideal conditions, as presented by a lot of researchers [41,48,183,185,187,188].

Other autonomous mechanisms, such as micro- and macro-capsules containing polymeric healing agents, may be independent of water availability; however, one-component polyurethane precursors need humidity to polymerize [36,162]. Any damage capable of disrupting the encapsulation system triggers the healing. It makes the method more reliable in terms of closure due to the occurrence of the cracking event, though it depends on a series of other variables such as the probability of cracks intercepting the capsules, and hence their distribution, the effectiveness of the healing agent in coming out of the capsule, type of shell and thickness, etc.

3.2 Healing of cracks considering the crack stress state and stability of the crack over time

In the broadest sense, the durability of a (construction) material has to be regarded as its ability to retain its intended performance over time under the anticipated structural service scenario. This does include not only the (aggressive) environmental conditions as discussed above but also a dedicated and careful consideration of the stress state in the structural element. Hence, the presence of “through crack” stress states has to be considered as well as their effects on the reliability and repeatability of the healing capacity.

In most reinforced concrete structural elements, tensile through crack stress states hold after the formation of a crack because of the cohesive nature of concrete fracture and the interaction with the reinforcement. These tensile through crack stress states can be either sustained throughout the intended lifespan or cycle between a minimum and a maximum value, with the amplitude of the variation and its frequency likely affecting the reliability and repeatability of the healing performance. On the other hand, and specifically in precast structural elements which during their service life go through different transient situations, tensile stress states in previously cracked regions may turn into compressive ones, with the same possibility of being sustained or cycling within a certain range as mentioned above.

Through-crack compressive stresses positively affect healing since the stress partially closes the crack and helps the crack-surface binding by the healing products. This condition might arise in precast columns or tunnel segments, which could crack during transport while experiencing axial or circumferential compressive stress states in their final in-service configuration. Ferrara et al. [189,190] investigated the effects of through-crack compressive stress on the healing capacity of notched beam specimens of different fibre-reinforced concrete mixes, either autogenous or stimulated by crystalline admixture and latex polymer, confirming the statements described above. On the other hand, through-crack tensile stresses have an adverse effect on healing. Özbay et al. [191] investigated the effects of sustained flexural loading in Engineered Cementitious Composites (ECCs), which is likely to widen the pre-induced cracks, on the effectiveness of the healing process. A slight decrease in strength recovery was found in specimens experiencing sustained flexural load compared to unloaded specimens [192]. The healing mechanism can become ineffective if the crack is active, so further experiencing cyclic through-crack stress states.

For example, continuous opening and closing of cracks can occur in concrete bridge elements under traffic loads or restrained elements, even simply experiencing thermal (day/night) cycles. The concern regarding the performance of self-healing materials in these conditions is due to the constant movement that can damage the healing products or detach them from the crack walls [31]. Depending on the self-healing mechanism, it may also be challenging for the healing products to form under these dynamic conditions. The mechanical properties of the healing agents are very relevant in this case since they influence the reoccurrence of damage [42]. If the material formed during healing is less resistant than the cementitious matrix, cracks tend to reoccur at the same site as the initial damage. If healing materials with greater resistance are applied, it ensures that posterior damage occurs at another point, ensuring the availability of the healing agent. For active cracks, the performance after healing also depends on the elasticity of the healing agent. If flexible enough, the healing agent can comply with the crack movement and the crack can be kept sealed, so repeated healing is unnecessary; if not flexible enough, repeated healing should be guaranteed [18]. Previous research has shown the effect of reloading by temperature variation after healing [162]. For cracks treated with water repellent agents (WRA), the absorption coefficient remained low even when the cracks grew upon reloading because the crack faces were still impregnated with WRA. At the same time, polyurethanes (PU) lost their bond with the crack surface, resulting in increased water absorption (Figure 5).

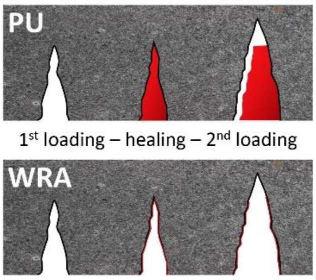


Figure 5. Effect of loading and reloading for self-healing concrete with PU (top) or WRA (bottom) [162].

In the case of healing agents providing the healing functionalities through expansive reactions, it has to be guaranteed that the volume variations induced by these reactions do not exceed the tensile deformation capacity of the concrete [42], otherwise, more cracks will likely appear. As an illustration, Sisomphon et al. [171] showed the excessive ettringite formation in concrete with expansive self-healing agents causing micro-cracking due to expansion.

Repeatability may be a concern in case of autogenous healing since the necessary healing materials are consumed over time by normal hydration of the matrix or by crack filling. Stimulated healing methods, such as the use of crystalline admixtures, have been found to be effective in facilitating long-term repeated healing for up to one year with re-loading after 1, 3 and 6 months, followed by healing regimes of 1 or 2 months under continuous water immersion by Cuenca et al. [27]. This has been attributed to osmotic migration of the active crystalline molecules throughout the concrete towards the freshly opened/reopened crack surfaces, where they can be consumed by the healing reactions. Similar results were obtained by Lo Monte and Ferrara [179] for autogenous healing of UHPC stimulated by crystalline admixtures. In the same framework, Prabhu et al. [193] developed a methodology to assess the effects of stimulated autogenous healing on the fatigue life of UHPCs. After pre-cracking the specimens and subjecting them to up to 700000 cycles between 10% and 80% of the pre-cracking load, specimens were healed for up to three months in water and then further reloaded for 100000 cycles after each scheduled healing period. With crack sealing of at least 80% after one month and up to 100%

after three months, the stimulated autogenous healing was instrumental for flexural stiffness recovery. It reduced the crack opening rate by as much as 18 times along the fatigue cycles, improving significantly the structure's fatigue life.

Feiteira et al. [194] assessed the performance of three encapsulated polyurethane (PU) healing agents with different viscosities under cyclic loading by reloading healed specimens in 3-point bending tests. The authors found that the lower viscosity PU (200 mPa.s) could heal cracks up to 300 µm and reduced the water uptake to the level of uncracked specimens. After reloading, the strain capacity supported by the healing agent was between 50% and 100%; hence, when increasing the crack width above 100% strain, the healing failed due to debonding of the PU from the crack wall. Nji and Li [195] developed a self-healing composite with the ability to heal repeatedly the damages by dispersing thermoplastic particles (copolyester) in a shape memory polymer matrix. Despite a healing efficiency of around 65% of the peak load being reached, it was only possible to apply the method when the samples were kept at 150°C for 20 min to activate the mechanism. Moreover, the fracture occurred after five cycles of loading. Botusharova [196] observed that a bacterium (*Sporosarcina ureae*) is able to go through cycles of self-healing multiple times in cemented sand columns without its activity being significantly affected. It was desirable not to provoke a repetition in the healing process at the same damage site due to the local consumption of nutrients. However, if there is a guarantee of survival in the matrix and the damage occurs later in another location where spores with activation capacity and nutritional availability still exist, it is expected that there will be MICP. Justo-Reinoso et al. [197] confirmed this by testing bacteria-based mortar samples and obtaining healing at later ages when new cracks form, but no repeated healing where the original crack was.

3.3 Stability of the healing functionality over time

The use of self-healing technologies in concrete is aimed at guaranteeing an increase in the durability of structures, extending their service life. These benefits can only be achieved when there is a guarantee of functionality stability over time in real conditions, representing the service scenario. Many studies have focused on the stability of the healing agent, considering only ideal environmental conditions [48,186], whereas few studies analysed the stability of self-healing mechanisms in real conditions [28,32,59,86,198–203]. Actually, a self-healing mechanism should be tailored to the scenario in which it will be employed. Knowing the limitations of each healing agent, it is possible to determine which methods and solutions can contribute to the survival of the agents. Some agents need protection to survive in extreme environments, for example, bacteria or reactive minerals, which must remain intact and without chemical or physical activity until damage occurs in the concrete. Survival is the main point to guarantee functioning over time since cracks may appear in concrete structures even after several years, upon which the healing agent should still be active to induce the self-healing operation.

Regarding (stimulated) autogenous healing, Ferrara et al. [177,178,204] reported that autogenous healing in HPFRCCs continued to be active even in specimens pre-cracked at about one year after casting and then exposed to the healing scenario. Cuenca et al. [27] verified that the effect of the crystalline admixture, employed as stimulator of the autogenous healing capacity in regular strength concrete, persisted up to one year upon repeated cycles of cracking and healing.

Regarding autonomous self-healing, extreme conditions can greatly influence the stability of healing agents. Polymeric healing agents are highly sensitive to high sunlight-induced temperatures, which can cause premature hardening inside capsules or variation in their viscosity [32,42]. Chemical reactions between the precursor and water are other phenomena observed by Gruyaert et al. [203] and resulted in curing of the precursor within the capsule. Moreover, the permeability of different types of capsule walls, or multilayer capsules, to resist the effects of water or oxygen present in the matrix should receive more research attention. This is important since permeable capsule walls can allow chemical interactions [205], leading to premature curing the healing agent and inactivating its self-healing property [206]. In the vascular-based approach, the continuity of the tube may be disturbed when the agent hardens inside a broken part of the tube, thus impairing its usability over time after rupture [42]. To solve these possible interruptions in the tubes, previous works suggested using an interconnected three-dimensional hollow network or “multi-walled” tubes consisting of a brittle outer tube and an inner flexible, permeable tube to preserve its continuity [207].

Bacteria are susceptible in their enzymatic and metabolic activity to changes in pH and temperature, the presence of salts or acids, or cold climates. The survival of bacteria inserted in cement matrices is a concern [48,208] and deserves more intense research efforts. However, there are indications that spores can survive in extreme conditions for a long time [197,200,209]. Studies on microbial concrete are mainly limited to short curing periods and crack formation at young ages (28 days or less) [183,186,188]. Limiting survival factors for the use of bacteria as healing agents are related to high pH values in concrete (pH ≈ 13), dense matrix (narrow pores), and unsuitable humidity conditions [17]. In order to guarantee the survival of these microorganisms within the cementitious matrix, protective measures can be taken, such as encapsulation [186,200] or biofilm formation by the organisms themselves [48,185,210]. The microbially induced $CaCO_3$ precipitation (MICP) using self-protected cultures such as activated compact denitrifying core (ACDC) granules is not only effective for crack healing at an early age but also closes cracks in mature specimens of 6 months [48]. Erşan et al. [48] noticed that with the ACDC, three days after the occurrence of the damage, the MICP process started. Wiktor and Jonkers [183] used bacterial spores (organic acids pathway) and nutrients embedded into expanded clay particles and mixed in mortar specimens. They were influential in closing cracks up to 460 µm formed at 56 days of age and healed for 100 days in water immersion. The authors noticed that bacterial crack healing started 20 days after water immersion, but it is essential to mention that the time for healing depends on several factors related to the healing conditions, such as the availability of water, bacteria-cell concentration and supply of nutrients. Justo-Reinoso et al. [197], using a similar encapsulation process, verified crack healing in new cracks formed in 22 month old samples after 8 weeks in partially submersed healing conditions. The needed level for factors such as pH, water activity, osmolarity, oxygen availability and temperature depends on the characteristics of each microorganism. For *Sporosarcina ureae*, a spore-forming, aerobic, ureolytic bacterium, encapsulated spores remained viable for at least six months, and multiple cycles of sporulation, encapsulation, and regeneration were possible in ideal (aqueous) conditions [200]. On the other hand, for *Sporosarcina pasteurii,* a highly ureolytic, aerobic bacterium, there was no guarantee of long-term survival [200].

3.4 Healing efficiency under different environmental conditions

There is much evidence from studies that high self-healing efficiency can be obtained; however, most of those studies consider the potential for self-healing under ideal conditions [23,26,211,212], while only a few consider a real or harsh environment [26,31,44,213]. In order to market self-healing technologies, the efficiency of healing agents must be guaranteed under the structural service scenarios, taking into account variables such as mix design, time of cracking, through-crack stress states, non-steadiness of cracks, and crack width [17,18,26]. Age of cracking and repeatability of the cracking and healing scenario relate to the stability and repeatability of the healing efficiency over time (section 3.3).

In addition to this guarantee, there is a need to guide the users on which healing agents should be selected to obtain the best performance in each possible real application situation. If self-healing efficiency is guaranteed, durability can be significantly improved even in contact with aggressive substances. Table 2 gives an overview (non-exhaustive) of self-healing agents studied in main aggressive scenarios so far. They are subdivided according to the mechanism as (1) autogenous or stimulated autogenous healing, (2) autonomous healing, and (3) bacteria-based self-healing concrete [17]. In the table, tests on self-healing agents in paste, mortar and concrete are all considered without distinction. Still, it should be noted that upscaling from mortar to concrete while maintaining the healing efficiency is a significant challenge. When the concentration of the healing additive is kept constant relative to the cement weight, the move from mortar to concrete results in a significant dilution of the additives. However, when keeping the same dosage in proportion to the total volume, an unacceptable strength decrease and high costs due to high healing agent dosage may result [17]. It is clear that most of the research in aggressive environments has focused on (stimulated) autogenous healing and exposure to chlorides and marine environment. So a noticeable knowledge gap exists regarding the performance of different types of self-healing materials in several aggressive scenarios.

Table 2. Overview of the research on different self-healing mechanisms in aggressive scenarios.

Main Aggressive scenarios	**Self-healing mechanism**		
	(Stimulated) autogenous healing	**Autonomous healing**	**Bacteria-based healing**
Chloride attack	**PC** [90,214] **SAP** [215] **SCMs** - Blast furnace slag [216] - Fly ash [26,217] - Silica fume [26] **Crystalline admixture** [26,28,44,166,218–221] **MgO expansive agent** [218] **Fibres** - UHPFRC [219,222] - ECC [217,223]	**Polyurethane** [78,224–228] **Silver alginate hydrogel capsules** [43] **Macrocapsules filled with water repellent agent** [46]	**ACDC bacteria** [210,229]
Carbonation	**Calcium nitrate** [230]		***Bacillus cohnii* bacteria** [230]
Sulphate attack	**SCMs** - Fly ash [231] - Blast furnace slag [231] **Fibres** [231] - ECC [232]	**Isophorone diisocyanate microcapsules** [233]	
Combined attack (marine environment)	**PC** [234] **SCMs** - Silica fume [235] - Blast furnace slag [213,236] - Fly ash [235] **Organic chelation agents** [237–243] **Crystalline admixture** [59,106,203,221,244–246] **Granules of $Ca(OH)_2$ and Na_2SO_4**[236] **Cellulose nanocrystals** [59,203,244,247] **Fibres** - ECC [232] - HPFRC [59,203,244,247] - Alumina nanofibers [59,203,244,247] - Cellulose nano-fibrils [59,203,244]		***Sporosarcina halophila* bacteria** [248,249]
Freeze-thaw attack with de-icing salts	**SCMs** - Blast furnace slag [250] - Fly ash [250] **Fibres** - ECC [251,252] **Crystalline admixture** [250]	**Water repellent agent** [253] **Sodium silicate** [253]	***Bacillus cohnii* bacteria** [254]
Freeze-thaw attack without de-icing salts	**Crystalline admixture** [237] **Fibres** - ECC [252,255]	**Toluene-di-isocyanate microcapsules** [256]	
Water pressure	**PC** [160] **SAP** [257] **Crystalline admixture** [25,172] **Fibres** - ECC [155,258]		**ACDC bacteria** [184]
High/low temperature and temperature variation	**PC** [259,260]	**Double-walled microcapsules** [261] **(PMMA-MA) capsules filled with MgO** [262] **Toluene-di-isocyanate microcapsules** [263] **Polyurethane** [32,162,264] **Sodium silicate** [201,205,265] **Water repellent agent** [162,264] **Shape memory polymers** [195,266] **Epoxy resin** [267]	**MICP** [196] **Bacillus cohnii bacteria** [268] **Psychrotrophic species** (Psy39) [268] **Bacillus halmapalus bacteria** [269,270]

Note: PC: Portland Cement; SAP: Superabsorbent Polymers; SCMs: Supplementary Cementing Materials; MgO: Magnesium Oxide; UHPFRC: Ultra-High-Performance Fibre-Reinforced Concrete; ECC: Engineered Cementitious Composite; ACDC: Activated Compact Denitrifying Core; MICP: Microbially Induced Calcium Carbonate Precipitation

3.4.1 Chloride attack

Research on the performance of self-healing concrete in chloride-rich environment has mainly considered the capacity of the cracks to heal. Recently, some studies also dealt with improving the durability indicators, such as the apparent chloride diffusion coefficient. However, studies that quantify the effect of self-healing on corrosion initiation of the reinforcement are scarce.

Autogenous sealing of micro-cracks was observed by Gowripalan et al. [90] in chloride-polluted environments. Darquennes et al. [216] performed chloride migration tests on self-healed mortars and claimed that, although blast furnace slag as SCM had a great impact on the self-healing process, crack width played a more significant role than material characteristics. Borg et al. [26] studied the sealing of cracks in mortars, including pulverized fly ash (PFA), silica fume (SF) and CA, when the specimens were exposed to continuous immersion and wet/dry cycles in distilled water, 33 and 165 g/L sodium chloride solution. The results were limited to measuring crack sealing capacity by image analysis of micrographs, with no tests to quantify the improvement of durability properties. Using PFA up to 20% improved the sealing of cracks below 150 μm. The addition of 15% SF allowed sealing crack widths between 150 and 300 μm. CA performed very well in most exposure conditions and for crack widths up to 300 μm. Both in submerged and wet/dry conditions, there were better self-healing results in chloride solution than in water [26].

The ability of crystalline admixture (CA) for self-healing in chloride environment has been investigated more recently [26,44,220]. Closure of cracks smaller than 300 μm was reached when submerged in chloride solution. For wet/dry cycles, cracks of the same width were also closed, but they took longer [26,44]. Due to its high hydrophilic character, CA improved the sealing of cracks up to 300 μm in every exposure condition. The higher the chloride content, the faster and better the crack healing with CA occurred. More recently, Cuenca et al. [219] studied the effect of autogenous healing and CA as a healing agent on the chloride ingress of cracked ultra-high performance fibre reinforced concrete (UHPFRC). Cracks with a width of 100 ± 50 μm were generated, and the cracked samples were immersed in 3% NaCl solution for 1, 3, 6 and 12 months. The results showed that the healing of the cracks postponed the ingress of chloride perpendicular to the crack and the healing efficiency of CA increased with longer exposure time. Abro et al. [220] studied the effect of a CA system consisting of Na_2CO_3 and organic Ca^{2+} together with zeolite (called SH-A) and the same CA with calcium sulfoaluminate (CSA) as an expansive agent and bentonite as a swelling agent (called SH-B). They adapted the ASTM C1202 to apply different voltages and calculated migration coefficients in mortar with 100 and 300 μm crack widths. The specimens were, after crack creation, subjected to two healing regimes submerged in water for 28 and 56 days. For SH-A, the components react with each other to produce $CaCO_3$ and fill the cracks, while for SH-B, the CSA incorporated as an expansive admixture creates ettringite and bentonite swells by absorbing water to fill the crack space. Specimens with 100 μm wide cracks after 56 days of healing had almost the same diffusion coefficient as the uncracked specimens, regardless of the mixture. For wider crack widths, concrete containing SH-B showed a greater reduction in coefficient than SH-A.

Other studies considering self-healing agents with polymethyl methacrylate or glass capsules as shells filled with water-repellent agent revealed that the resistance of cracked concrete against chloride could be increased [46]. Xiong et al. [43] presented a novel capsule-based self-recovery system with a chloride ion trigger, which is functionalized via a smart response to chloride ions. A chloride concentration of 0.1 wt% is already sufficient to disintegrate the capsules of silver alginate hydrogel, releasing the activated core materials for self-recovery [43]. Van Belleghem et al. [225] used macro-encapsulated polyurethane to heal the cracks in concrete exposed to 165 g/L of sodium chloride solution. The chloride concentration of the healed specimens at the anodic surface of the reinforcement was reduced by about 75% compared with cracked specimens without polyurethane [227].

Concerning the role of superabsorbent polymers (SAP) in self-healing, Van Mullem et al. [215] verified the chloride ingress in mortar samples with a crack width of 150 μm with and without SAPs. The healing regime of wet/dry cyles was performed for 28 days before submersion of the specimens in a chloride solution with 150 g/L of sodium chloride. The use of SAP stimulated autogenous healing, and there was a considerable reduction in chloride ingress in the first week of exposure. However, healed SAP specimens had a similar chloride ingress as healed reference specimens after exposure to aggressive media for five weeks.

As mentioned above, studies on the actual effect of self-healing on corrosion of reinforcement in chloride-containing environments are rare. Van Belleghem et al. [227] performed corrosion monitoring on concrete prisms 120x120x500 mm^3 containing a steel reinforcement configuration with spatially separated anode and cathode, allowing measurement of the macro-cell corrosion current. A flexural crack of 300 µm was self-healed with macro-encapsulated polyurethane and exposed to wet-dry cycles with a 33 g/L NaCl solution. Figure 6 shows corrosion initiation in cracked samples after 6 to 10 weeks of exposure, while in self-healing samples with low viscosity polyurethane (PU_LV) corrosion did not initiate over the 26 weeks of testing.

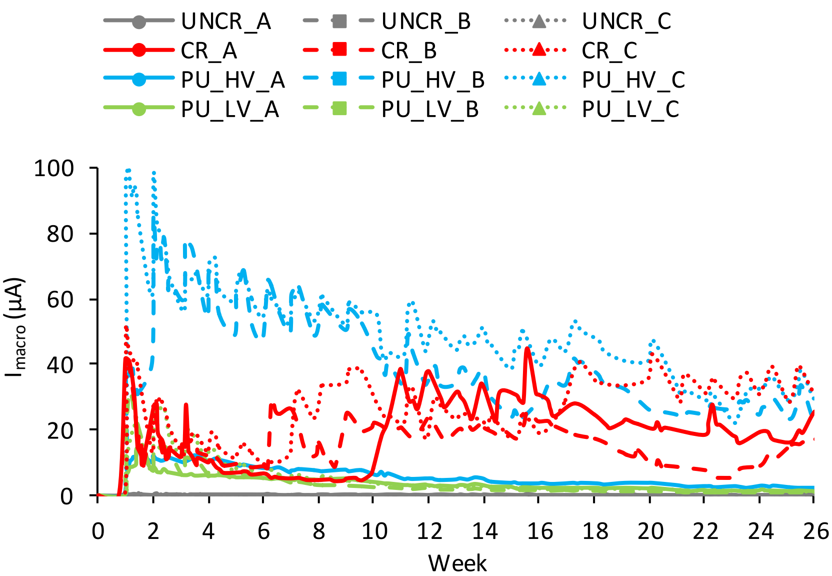


Figure 6. Electrochemical corrosion parameters of uncracked samples (UNCR), cracked samples (CR), samples healed with high viscosity polyurethane (PU_HV) and samples healed with low viscosity polyurethane (PU_LV). [227] *Copyright 2018, Elsevier.*

Corrosion inhibition by nitrite produced by nitrate-reducing bacteria as self-healing agents can only be proven by actual corrosion tests. Erşan et al. [229] were the first to prove the inhibitory effect of the microbially produced nitrite on steel plates immersed in a chloride-containing solution. Then, ACDC microbial culture (0.5% wt/wt cement), was added to a cementitious matrix together with $Ca(NO_3)_2$ (3% wt/wt cement) and $Ca(HCOO)_2$ (2% wt/wt cement) [210]. Mortar specimens contained 300 µm artificial cracks, running up to a central steel rebar. The open circuit potential (OCP) was measured for 120 days and compared with the limit OCP value (–250 mV vs standard hydrogen electrode), while the cracked surfaces were immersed in 0.5 M Cl^- solution. For the cracked plain mortar (Figure 7), rebar corrosion initiated after 16 days of exposure to Cl^- solution (OCP drop below –250 mV), whereas the ACDC-containing specimens gave proof of corrosion inhibition by the produced nitrite during the first 28 days when the cracks had not yet been completely healed. The measured mass losses of the steel reinforcement pointed at similar rebar protection as for uncracked mortar.

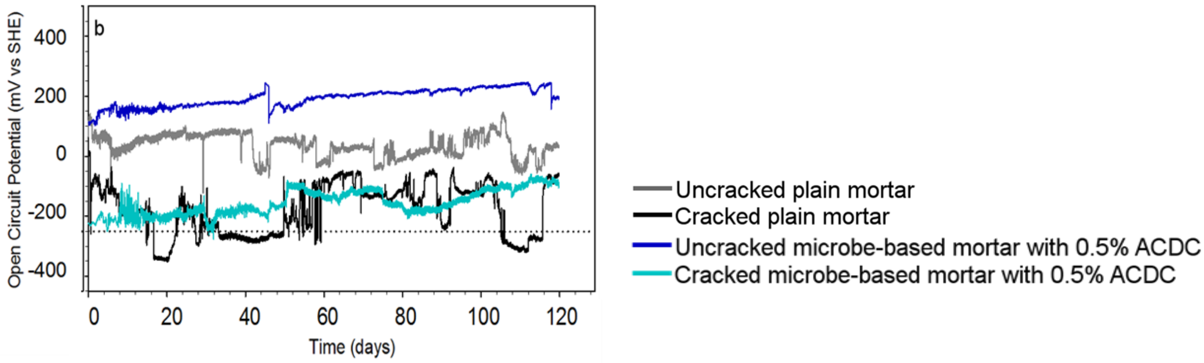


Figure 7. Open circuit potential (OCP) versus standard hydrogen electrode (SHE) for uncracked and cracked plain mortar and self-healing mortar with denitrifying ACDC. The dotted line shows the limit OCP [210]*. Copyright 2018, Elsevier.*

3.4.2 Carbonation

Typically, autogenous healing of cracks in neutral environments is based on two main mechanisms, 1) further hydration of unhydrated cement particles sealing the crack and 2) interaction of the hydrated cement paste phases (calcium hydroxide and C-S-H) with the CO_2 dissolved in water, forming calcium carbonate that seals the crack. These reactions occur when the concrete is under immersion or in high moisture environments [21,271,272]. Most autogenous healing processes lead to the precipitation of calcium carbonate in the crack; however, the concrete matrix generally has not suffered from the carbonation in these cases.

There are a limited number of studies related to the self-healing of carbonated concrete or the effect of self-healing on concrete carbonation. Alahmad et al. [95] showed no carbonation along cracks less than 9 µm wide due to the autogenous healing of concrete leading to the crack closure. Tan et al. [230] studied the effect of carbonation on crack self-healing. The authors searched for two different ways of making more calcium ions available for the healing process in mortars: 1) adding calcium directly in the form of a solution of calcium nitrate to the mix design, and 2) adding encapsulated calcium in combination with a bacteria-based self-healing system. They showed that self-healing of non-carbonated mortar was more efficient than for carbonated mortar since there were more calcium ions available. An encapsulated source of calcium ions aided the self-healing of carbonated mortars. In carbonated bacteria-based mortars, calcium carbonate precipitated on the surface of the bacterial biofilm, and this led to the closing of the crack.

3.4.3 Sulphate attack

Only a few studies related to self-healing processes in sulphate-containing environments can be found [231–233], probably due to the fact that sulphate attack mainly occurs combined with chlorides in marine environments [86]. He et al. [245] focused on an in-house developed crystalline admixture and reported an increased sulphate resistance of uncracked specimens considering wet/dry cycles with 5% of sodium sulphate solution. Dobrovolski et al. [273] studied internal sulphate attack by using pyrite, which could release sulphate ions upon oxidation, and investigated the effect of using CA. Both did, however, not analyse the effect for cracked or self-healed samples. Nevertheless, using CA combined with sulphate can contribute to gypsum, ettringite and aragonite crystals formed around the cracks. Liu et al. [232] induced multiple tensile cracks up to around 150 µm in ECC. The specimens were then immersed directly in a solution containing 5% of sodium sulphate. The authors reported that ECC tends to heal faster and more completely in sulphate solutions than in water, closing the cracks after 120 days, probably due to the ettringite/gypsum formation within the cracks [232].

Hung and Hung [231] considered a sodium sulphate solution as a medium to promote autogenous healing in a type of innovative fibre-reinforced material with cement replaced by ground granulated blast furnace slag (GGBS) or fly ash (FA). Cracks between 60 µm and 120 µm showed a two times higher closure in sulphate solution than in water after 28 days. GGBS concrete had a higher crack reduction ratio than FA concrete for crack widths below

100 μm, and the presence of fibres allowed to regain the strength and stiffness due to the precipitation of the healing product on the fibres. The authors verified the formation of calcium carbonate, calcium silicate hydrates, and ettringite mainly deposited within the cracks.

Regarding encapsulation systems, Du et al. [233] considered microcapsules with isophorone diisocyanate (IPDI) as the healing agent and paraffin wax, polyethylene wax, and nano-silica as shells to promote sulphate resistance by healing cracks formed due to sulphate attack. In theory, the capsules should break during crack propagation, releasing the healing agent and filling the cracks. However, the evidence was insufficient to prove that this happened, although an increase in sulphate resistance in the mortar was noticed.

3.4.4 Combined attack (including marine environment)

Most of the self-healing studies that contemplate combined attack consider performance in marine environment (sulphate-chloride attack). However, other combined attack mechanisms can also be found in the literature as the effect of pressurized water and sustained load on engineered cementitious composite (ECC) [258]. In [258], the authors verified that cracks of 100 μm healed entirely after ten days under continuous water pressure and sustained load, blocking the leakage, and the main products formed were $CaCO_3$ and C-S-H/CH.

Recently, Cuenca et al. [59,203] and Lo Monte and Ferrara [244] have investigated the crack self-healing capacity of UHPC made with different types of cement (CEM I and CEM III) with CA, in case also associated with specific nano-additions (alumina nanofibres and cellulose nanocrystals and nanofibrils), when exposed to natural geothermal water containing both chlorides and sulphates. Both permanent immersion and exposure to wet/dry cycles were considered, and effects of re-cracking after scheduled healing times, as long as six months, were investigated. Single localized cracks opened up to 150 μm, and multiple cracking states with single cracks not wider than 100 μm were studied. While the self-healing performance of UHPC made with CEM III was generally better than with CEM I, the differences tended to fade upon multiple re-cracking and longer exposure times. Effects of nanoparticles were evident in enhancing the crack-width control capacity of the material, guaranteeing single cracks not wider than a few tens of microns for the same level of deformation, and hence guaranteeing a better healing performance [247]. For the same UHPCs and aggressive scenario, Giménez et al. [247] have developed a tailored test set-up to simulate interaction with continuously flowing water as it occurs, e.g. in cooling towers in geothermal power plants. They found that while the continuously flowing water may exacerbate the effects of previously induced cracking, it resulted in partial healing after two years of exposure, considering cracks up to 50 μm opening. The inclusion of nano-additions resulted in a calcium-rich protective layer, also affecting the nature of the crack healing precipitates (less presence of ettringite in the inner part of the cracks because of lower penetration of sulphates).

In marine conditions, the deterioration of concrete is also caused by multi-ion attack, the main ions being chlorides (Cl^-) and sulphates (SO_4^{2-}), both competing for binding with the C_3A of the cement [86]. According to several authors, autogenous healing is improved under seawater immersion, especially when cement is partly replaced by blast-furnace slag (BFS) and fly ash [274–276]. On the other hand, Palin et al. [277] showed that after 56 days, BFS specimens could heal cracks completely when the widths were up to 104 μm in seawater and 408 μm in freshwater. Cracks also healed in PC specimens when their openings were 592 μm and 168 μm in seawater and freshwater, respectively. The researchers claimed that the healing process in these cracked specimens was related to the availability of calcium hydroxide in mortars and specific ions in seawater. Due to the presence of magnesium ions in the seawater, the combined action may contribute to brucite precipitation, causing additional sealing of the cracks [213]. Besides, Mohammed et al. [81] observed that products like ettringite, calcite and brucite sealed small microcracks when exposed to a marine environment. Danner et al. [235] investigated the mineralogy of the self-healing products formed in cracked marine concrete after 25 years of exposure. They confirmed the closure of cracks with widths smaller than 200 μm. While calcite formed in the outer part of the crack (0-5 mm in depth) followed by a brucite layer (5-30 mm depth), deeper into the crack, only ettringite was observed. Xue [221] studied two types of CA in concrete exposed to seawater and observed that the self-healing performance is strongly related to the type of CA combined with the environmental condition. The author produced cracks with a width of 60 to 70 µm in mortars to be analysed under immersion and wet-dry cycles. The wet-dry cycles increase the chloride ingress due to the capillary suction compared to the immersion. He also considered using MgO-cement to collaborate with autogenous healing in seawater and chloride solution. The seawater reduces the chloride ingress compared to a pure chloride solution because of the brucite precipitation in the cracks [246]. He et al. [106] applied a non-commercial crystalline admixture in mortar exposed to seawater and could heal completely cracks up to 350 µm, while in freshwater, the maximum crack width healed was 240 µm. They verified brucite and calcium carbonate as healing products in the cracks.

The healing agent used in seawater conditions must guarantee efficiency because of the combined attack. In the case of bacteria-based self-healing concrete, a bacterium must be chosen suitable to survive in seawater. Palin et al. [269] developed a bacteria-based self-healing cementitious composite for application in low-temperature (8°C) marine environments, showing almost complete healing of cracks; 95% healing capacity for cracks up to 400 µm in width and 93% for 600 µm wide cracks [269]. Erşan et al. [210] analysed the tolerance of a nitrate-reducing ACDC microbial culture to osmotic stress in the marine environment. The major influential parameter appeared to be the ionic strength (µ) of the solution rather than a change in specific ion concentrations. In typical seawater (µ ~ 0.7 and $[Cl^-]$ ~ 0.5 M) results revealed that ACDC culture showed adequate resistance [210]. Khan et al. [248] used spores of *Sporosarcina halophila* bacteria, calcium lactate and expanded perlite aggregate as the carrier in mortar subjected to submerged and tidal marine environment for 90 days. The presence of bacteria and availability of water and oxygen within the crack enhanced the aragonite and brucite formation along the entire crack depth, with a crack closure ratio of 50% considering cracks up to 800 µm.

The improvement of autogenous healing due to the use of blast furnace slag (BFS) is well known [274–276]. However, the reaction usually takes time, and in realistic aggressive environments, it might not prevent the aggressive agent from entering before the healing occurs. In this respect, the use of alkali-activators combined with BFS can bring benefits to improve healing. Kim et al. [236] tested granulated calcium hydroxide or sodium sulphate, enveloped in PVA film, as an activator for self-healing in mortars exposed to seawater. The authors identified complete closure of cracks smaller than 300 µm after 60 days of exposure when using calcium hydroxide as an activator; sodium sulphate was ineffective. Liu et al. [239,240] proposed using organic chelation agents to efficiently block cracks in a marine environment. The mechanism involves an innovative way to rearrange the crystals in the cement matrix related to the growth process and shape by means of their chelation activities [237,238,241]. Liu et al. [240] prepared cement pastes with 1.5% vs cement of tetrasodium ethylenediaminetetraacetic acid (EDTA-4Na), triethanolamine (TEA), or sodium hexametaphosphate (SHMP) and exposed them to seawater (submerged condition/wet-dry cycles). The TEA agent could heal completely cracks with a width of 400 µm after two days under immersion in seawater due to an increase in the OH^- concentration, inducing the rapid formation of brucite (Figure 8). For cracks of 800 µm, 56 days of exposure were necessary to reach a crack closure of 97%. Using 1% of TEA (by the mass of cement) closure of 400 µm wide cracks was obtained 20 times faster than for a control paste [239]. While for autogenous healing in seawater cracks up to 150 µm could close for both submerged and wet-dry cycles, cracks of 400 µm reached only 90% of closure in wet-dry cycles, and 60% under submerged condition [234]. Wu et al. [242] considered to include 5% by volume of CaO-$NaAlO_2$ in cement paste and exposed the samples to seawater for accelerating the healing process by binding chemically the ions present in this solution. They could heal cracks of 400 µm wide in 1 day of seawater immersion. However, there is no evidence how these additions can affect a concrete structure in the long-term with regard to performance and durability.

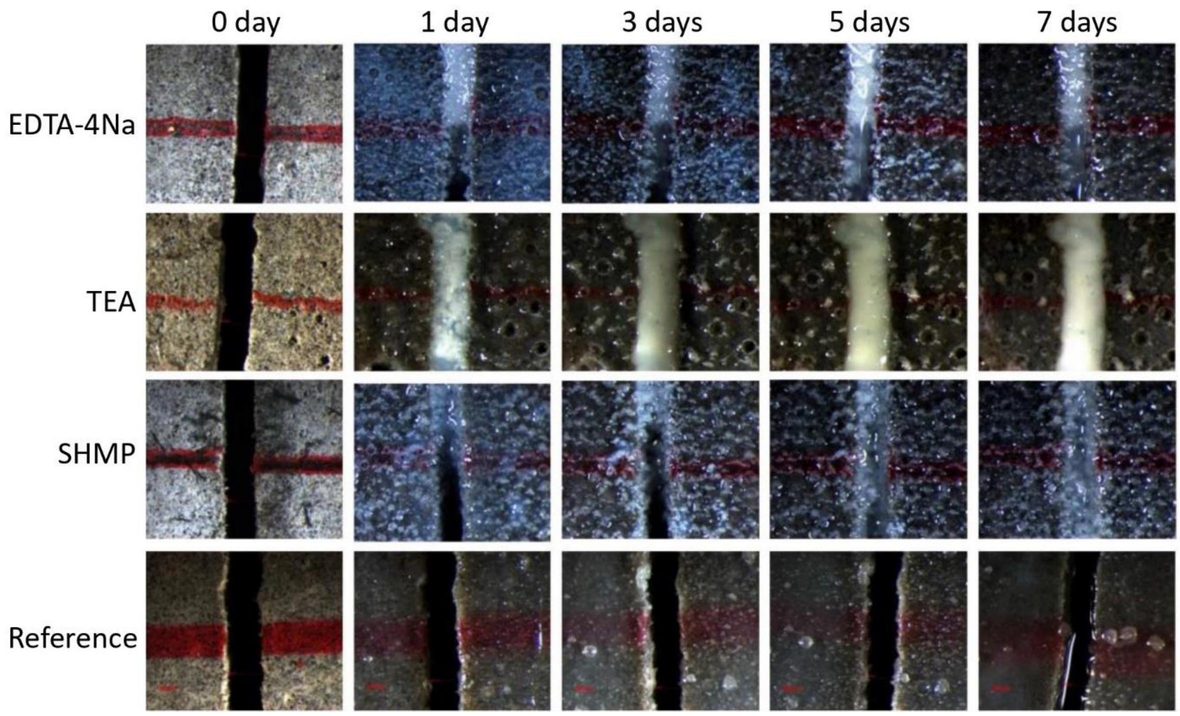


Figure 8. Crack closure by image analysis of self-healing organic chelation agents in cement paste with crack widths of 400 µm immersed in seawater [240]*, with permission from ASCE.*

3.4.5 Freeze-thaw attack with or without de-icing salts

In order to increase the durability of structures subjected to freeze-thaw (FT) and reduce internal damage, the most appropriate way is to reduce the possible moisture uptake. FT tests on cracked and self-healed specimens showed clearly that the deterioration was governed by the capacity of water penetration through cracks [278]. Self-healing mechanisms have been studied to a limited extent, but they can contribute to a large degree in guaranteeing the reduction of water entry through the closure of cracks. Most research related to this extreme condition shows the improvement by reducing matrix permeability or by methods to reduce the crack propagation [133,135,252,279,280]. Other studies are restricted to the use of products which increase the scaling resistance of the surface layer of concrete structures [133,135].

SAPs can be added to cementitious materials to increase the FT resistance [279] and induce self-sealing and self-healing effects [280]. SAPs create macropores that act in the same way as air voids created by an air entraining agent. These macropores also attract microcracks formation, stimulating multiple cracks of reduced width and contributing to self-healing [280–284]. Due to the addition of SAP, a much less pronounced decrease in the dynamic modulus was observed as a result of FT testing without de-icing salt [279]. However, published research does not include the analysis of cracked and healed SAP-containing concrete in this extreme condition but is limited to the increased frost-thaw resistance of uncracked matrices.

Suleiman and Nehdi [260] evaluated autogenous self-healing in mortar specimens under water submersion at a constant temperature of 19°C and in an environmental chamber under cyclic temperature from −10°C to 40°C and relative humidity in the range of 20% to 90% while monitoring during one year. The complete cycle was twelve days, four days at 20°C and 90 % RH, four days at -10°C and 20 % RH, and four days at 40°C and 60 % RH. They did not report significant healing in the cyclic temperature environment even for 30 µm crack width, while in submerged conditions, cracks with 300 µm width were closed entirely. However, one could comment that such accelerated tests might not allow sufficient time for healing to occur between the low temperature parts of the cycles. Jacobsen et al. [134] subjected concrete samples (binder containing 5% silica fume) to FT cycles to cause microcracking (1-10 µm width cracks). After damage, they completely submerged the samples in water where the specimens could heal autogenously; however, the samples could not recover their pristine performance according to resonance frequency and compressive strength tests. In another research also regarding autogenous healing, concrete that lost 50% of the dynamic modulus due to frost damage could almost completely recover after subsequent storage in water, somewhat varying with concrete composition and degree of deterioration [278].

However, in real situations, it is unlikely that structures will have ideal curing to guarantee the healing after the damage. Ma et al. [150] analysed concrete specimens subjected to FT cycles with de-icing salts according to the standard GB/T 50082–2009. Each freeze–thaw cycle lasted for nearly 4 h, the thawing time being longer than 1 h. The central temperature of the specimens was controlled subsequently at -18 ± 2 °C and 5 ± 2 °C. After cracking occurred due to the FT actions, they noticed no tendency of autogenous healing under the given exposure condition where the samples was kept in cold condition. Zhu et al. [252] noticed that ECC had weakened properties when submitted to FT conditions, and in the presence of salts, the self-healing degree was worse than in the condition with only water**.** In contrast, Şahmaran and Li [251] reported that ECC has the capacity to heal sufficiently under freezing and thawing cycles in the presence of salt solutions to restore nearly the original stiffness because of the multiple cracks with a narrow width. Hooshmand et al. [255] studied large-scale panels made with ECC (with blast furnace slag or fly ash) and PC for seven months in a natural environment that suffered FT cycles during certain periods. They noticed the greater presence of C-S-H compared to calcium carbonate in cracks healed in the ECC made with fly ash compared to the one made with slag. The authors believe that this might be the reason for the higher resistance of the healing products in the fly ash ECC mixes to the harsh natural conditions. In this case, the fly ash led to a better self-healing performance of cracks formed, while the ECC with slag had higher crack formation exceeding the recommended crack width limit (100 µm) as result of higher shrinkage strains.

Zha et al. [237] analysed a novel CA as an ion chelating agent in mortars after 100 FT cycles. The specimens were subjected to a further healing regime of 28 days submerged in water after FT. The CA mortars showed a compressive strength recovery of 51.8%, and cracks with a width of 320 µm could be healed. Needle-like crystals were observed in the pores and micro-cracks due to the freezing, with calcium carbonate, C-S-H and ettringite being identified as the main components. Wang et al. [250] investigated a non-commercial CA with blast furnace slag or fly ash. First, mortar specimens were subjected to 100 FT cycles with 3 wt% NaCl. Subsequently, the damaged specimens were subjected to a healing regime (water immersion) for 28 days. The authors verified the healing ability after exposure by compressive strength tests. The mortar made with CA and blast furnace slag recovered about 22% of the strength and reduced about 27% of harmful pores (larger than 0.1 mm).

Concerning autonomous self-healing mechanisms, Du et al. [256] used toluene-di-isocyanate (TDI) microcapsules in concrete specimens subjected to 100 FT cycles to verify the ability to heal cracks after FT. After exposure, the specimens were inserted in a healing regime of 7 days under water immersion. The authors believe that the damage caused by crack propagation was able to break the capsules to release the TDI within the microcracks. The TDI might react with the water, healing the cracks and improving the FT resistance. Cappellesso et al. [254] considered a self-healing bacterial concrete exposed to FT associated with chlorides. The *Bacillus cohnii* bacteria increased the performance of the concrete reducing the scaling by 90% and chloride ingress by 46% under FT compared to a reference concrete without bacteria. The addition of the bacteria had an air-entraining effect that might explain the improvements reached. However, it could not block the chloride ingress through a pre-defined crack of 170 ± 50 µm healed during 28 days of water immersion before exposure. The healing products formed by the bacteria before exposure to freezing temperatures were damaged by scaling due to freezing during the subsequent exposure period. The same authors [253] also considered two types of healing agents (water repellent agent and sodium silicate), injected them in cracks of 100 µm wide, and subjected them to FT scaling.

The sodium silicate after a healing regime of 14 days in a layer of water could prevent the chloride ingress through the crack after 56 FT cycles with 3 wt% NaCl.

3.4.6 Water pressure

Many studies related to self-healing mechanisms assess the influence of water pressure through water permeability methods [25,36,285–293,172,184,194,212,213,224,225,257]. Figure 9 shows some examples of test methods used to verify the self-healing performance under low or high water pressure. However, most of these studies aim to quantify the crack closure by the reduction in permeability to water under a certain pressure, which usually is smaller than 0.005 MPa (0.05 bar), without aiming to assess the ability of the healing agent to face the hydraulic pressure exerted on the healing products [25,172,257]. Gruyaert et al. [257] analysed mortars made with SAP to investigate the resistance of the healed specimens to a high water pressure of 0.2 MPa. The authors could verify the sealing efficiency by water flow tests on mortar made with SAP having 150 µm of crack width and healed in wet-dry cycles for 28 days.

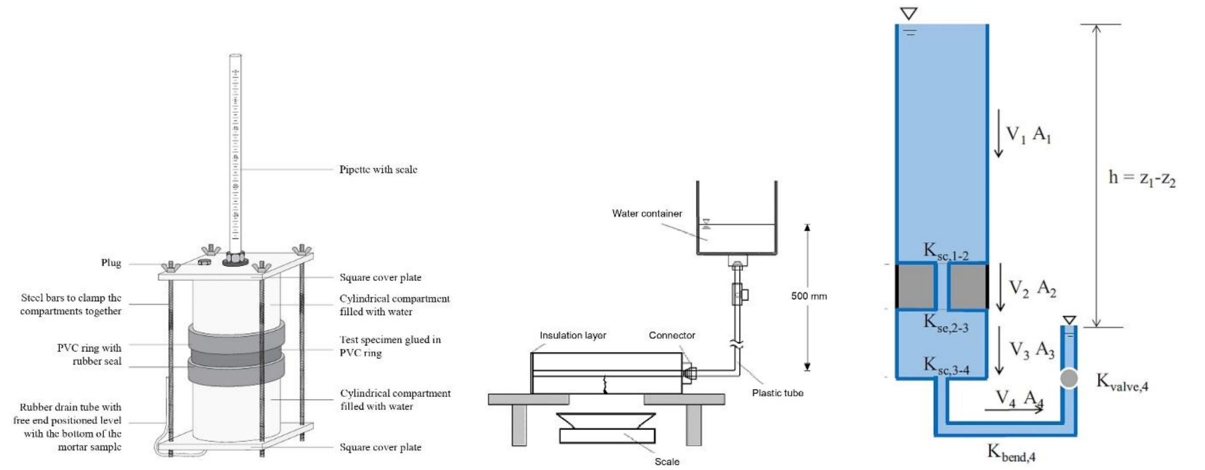


Figure 9. Set-ups for water permeability test under low/high pressure: (a) Aldea set-up for water permeability [285] *Copyright 2011, Elsevier., (b) HEALCON water permeability test* [289] *Copyright 2019, Elsevier, (c) Korean water permeability test* [294]*. Copyright 2017, Elsevier.*

CA have been applied in real structures with the aim of reducing the permeability of concrete essentially in tanks, containment walls in contact with groundwater, tunnels and reservoirs, i.e. structures where water is acting with a pressure gradient. Borg et al. [26] called attention to the fact that structures under water pressure present an ideal condition for the crystalline admixture because of the huge availability of water. Nonetheless, research is needed to determine the hydraulic pressure threshold that can be supported by the formed healing products and the effect of hydraulic pressure on the continuation of the cracking process, which can increase the damage and leakage in these structures. Regarding autonomous healing, Erşan et al. [184] measured the healing performance of biomortar with ACDC under 0.01 MPa of pressure in a water permeability test. The cracks of 400 µm showed 80% decrease in the flow rate after 28 days of healing in water immersion.

ECC has shown high potential to withstand extreme conditions, mainly because it contains fibres, contributing to the reduction in the crack widths. Hooshmand et al. [155] verified the efficiency of ECC, pre-cracked in direct tension up to two different crack levels and healing under 0.048 MPa water pressure, in short-term (up to 30 hours - Figure 10a) and in long-term (up to 10 days - Figure 10b). The authors showed the potential of the investigated high-performance cementitious composites to heal better under water pressure, as compared to conventional concrete, rightly due to the capacity to spread the localized damage into multiple cracks. Closure by autogenous self-healing in less than a week was observed for the 50 µm wide cracks (‘South’ in Figure 10b), whereas, for the crack with a width of 100 µm (‘North’ in Figure 10b), ten days were needed to quash the leakage

[155]. SEM-EDX analyses showed that the cracks of ECC were filled with higher quantities of dense self-healing products ($CaCO_3$ and C–S–H / CH) compared to conventional concrete.

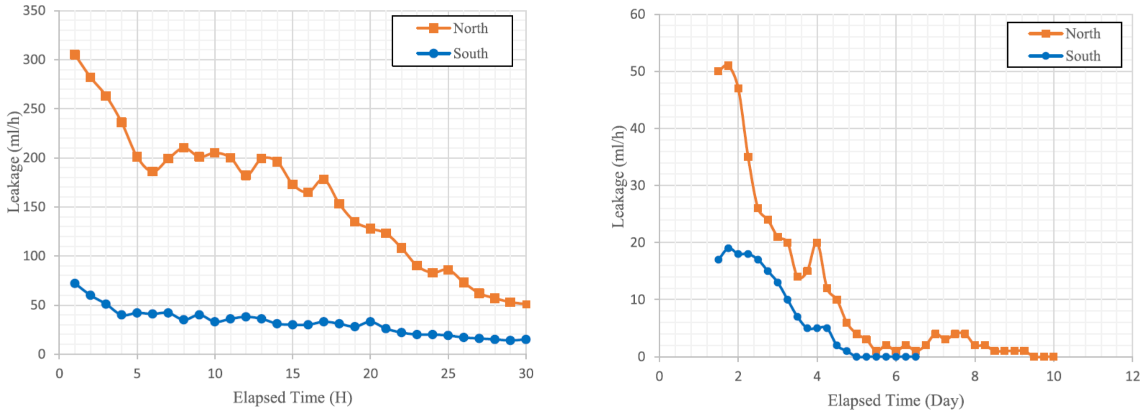


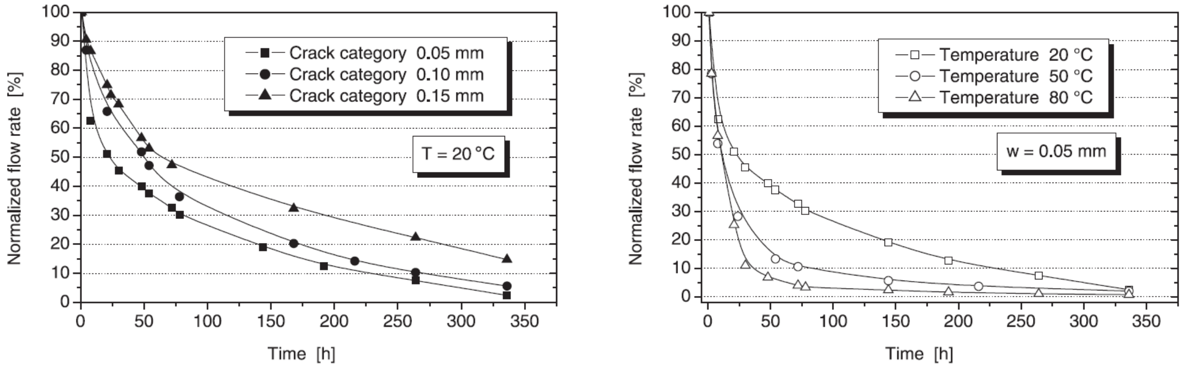
Figure 10. Outcomes of leakage rate versus time in ECC under water pressure: (a) short-term self-healing in hours, (b) long-term self-healing in days. North represents cracks with a width of 100 µm, while South represents 50 µm wide cracks [155] *Copyright 2021, Elsevier.*

3.4.7 High/low environmental temperatures and temperature variations

A few studies have been devoted to assessing the persistence of the healing capacity under or after exposure to a non-standard temperature, which can cause issues to the survival of the healing agent, whether it is a mineral, bacterium or an encapsulated polymer.

In bacteria-based self-healing concrete, the use in sporulated form brings benefits for application at high temperatures, given their resistance to survive in these conditions [200]. Botusharova [196] demonstrated that the MICP biosealant used was thermally stable at temperatures up to 840°C. Contrarily, ureolytic activity is mainly maintained at temperatures ranging from 4°C to 60°C [196]. Palin et al. [269] applied a bacteria-based self-healing cementitious material containing *Bacillus halmapalus* spores at a low temperature of 8°C submerged in seawater. They obtained a healing efficiency of 95% for 400 µm crack width and 93% for 600 µm crack width after 56 days. Paine [295] called attention to the need to choose bacteria fit to the conditions, e.g. types that can withstand hot temperatures (thermophilic), low temperatures (psychrophilic), and/or presence of salts (halophilic). Skevi et al. [268] focused on the survival of bacteria-based self-healing agent at low temperature (7.5 ± 2°C) where a psychrotrophic species (Psy39) and *Bacillus cohnii* were used. Mortars with cracks between 400-500 µm made at 28 days age were subjected to a semi-submerged condition at 7.5 ± 2°C and 20°C during more than 28 days. While *Bacillus cohnii* bacteria have better performance at 20°C, the Psy39 reached the greatest efficiency for both temperatures.

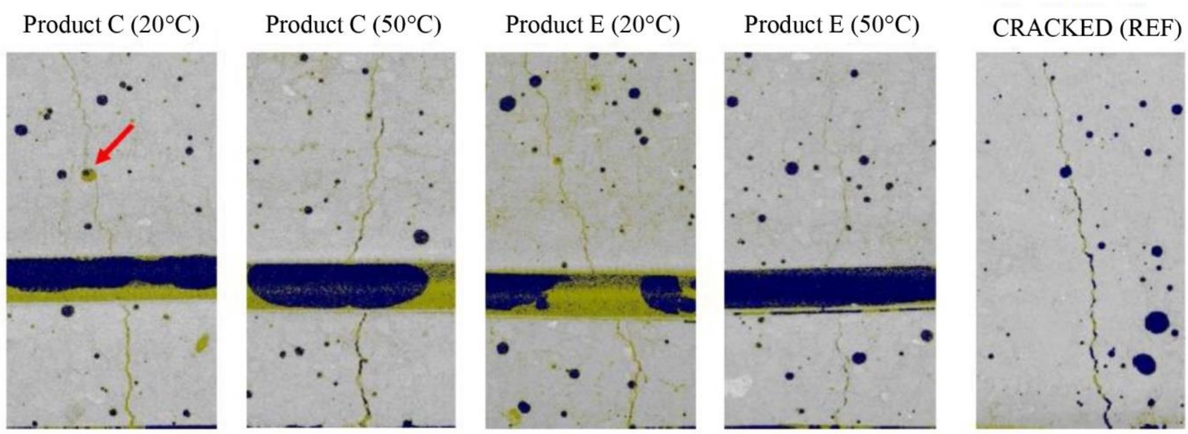
Reinhardt and Jooss [259] assessed autogenous self-healing under water in a high-performance concrete at 20, 50 and 80°C, relating it to variation of crack width. A clear tendency for a faster self-healing process in case of an increase in temperature could be recognized and is presented in Figure 11.



(a) (b)

Figure 11. Decrease of the normalized flow rate because of the self-healed cracks for HPC at a pressure gradient of 1 MPa/m (a) with various crack widths and a temperature of 20 °C, (b) with various temperatures and crack width of 50 µm. [259] *Copyright 2003, Elsevier.*

Van Tittelboom et al. [264] assessed the efficiency of encapsulated polyurethane and water-repellent agent as healing agents in case of thermal crack formation in sandwich panels. Similar research by Gruyaert et al. [162] in a real-scale test evaluated self-healing mechanisms in the presence of thermal cracks of around 50 µm and 150 µm. Polyurethanes (PU) and water repellent agents embedded in glass macrocapsules were both able to reduce the permeability by healing the thermally induced cracks, but under reloading, only water repellent agents retained the crack watertight. The PU lost the bond with the crack wall increasing water absorption and showed moreover a high leakage out of the cracks due to the decrease in viscosity because of the high temperature (~55°C) [162]. Van Belleghem et al. [32] tested five different types of polyurethanes to verify the efficiency of self-healing for concrete exposed to high temperatures (50°C). They showed that for the specimens healed with Product E, a certain type of polyurethane, at 50°C, the glass macrocapsules were completely empty (Figure 12d). It is related to the reduction by around 85% of the polyurethane viscosity when the temperature was increased from 20°C to 50°C. The excessively low viscosity of the healing agent can impair the healing efficiency as the polymer may leak from the crack [32,162]. This indicates that tailoring of the healing agent's viscosity to the expected temperature conditions is a key issue in obtaining greater self-healing efficiency.



(a) (b) (c) (d) (e)

Figure 12. X-ray µCT cross sections showing the crack intersecting an embedded macro-capsule for a specimen healed with Product C at (a) 20°C and (b) 50°C; a specimen healed with Product E at (c) 20°C and (d) 50°C; and (e) a specimen with an untreated crack, with permission from ASCE [32].

Regarding self-healing systems with microcapsules, Mostavi et al. [261] showed that their double-walled microcapsules provide enhanced durability at high temperatures compared with single-walled microcapsules preserving a suitable interfacial bonding. Du et al. [263] presented different shell materials which were analysed under different temperatures (from 10°C to 60°C), containing toluene-di-isocyanate as core material. The authors were concerned regarding the thermoplastic materials commonly used to produce shells, such as paraffin, polyethylene, or polypropylene. For instance, paraffin has a melting point of around 60°C; thus, at high temperatures, the shell could be destroyed before being broken by the cracking process. Then the healing agent inside the microcapsules may not flow out to heal the crack at the right time, or the healing agent efficiency could be affected. Kanellopoulos et al. [205] indicated that microcapsules with sodium silicate solution as a core and gelatine-acacia gum as a shell were stable under thermal effects up to 190°C. Mao et al. [201,265] demonstrated that sodium silicate microcapsule-based self-healing of oil well cement could present a good stability at the high temperature of 80°C. Ren et al. [262] applied innovative microcapsules with temperature-adaptive properties and analyzed how those capsules affect the concrete properties when the concrete mixing is developed at normal temperature (20°C) and high temperature (60°C). The capsules were made by polymethylmethacrylate-methacrylate (PMMA-MA) shells and magnesium oxide core. The authors verified 3% higher healing efficiency considering the high temperature due to the fact the PMMA-MA shell has a glass transition temperature of 40°C, so less premature rupture of the shell appears when mixing at 60°C.

Thermal effects can also be applied to help heal cracks. Shape memory polymers (SMP) act as tendons which contract upon heating to restore the original shape, creating a prestress in the element, closing the crack mechanically and, as such, enhancing the healing action [195,266]**.** Li and Neetles [296] compared two types of SMP, reaching almost 100% of recovery by thermomechanical analysis and closing completely the crack with no specification of the width. Nji and Li [195] considered an SMP with 6% of copolyester particles obtaining repeatability of 5 heating cycles. Teall et al. [266] made use of an SMP (polyethylene terephthalate [PET] filament) to close cracks of 750 µm by electrical heating activation and reached a crack closure of 85% in unreinforced beams (Figure 13), and 26-39% considering reinforced beams. They believe that the reinforcement can cause interference in the electrical activation. Nishiwaki et al. [267] produced a self-healing system for concrete that incorporates a heating pipe device filled with a repair agent, which was a one-component epoxy resin. With an increase in temperature around the crack, the organic film (ethylene vinyl acetate [EVA] polymer) that composes the pipe melts at 93°C allowing the healing agent to release within the crack.

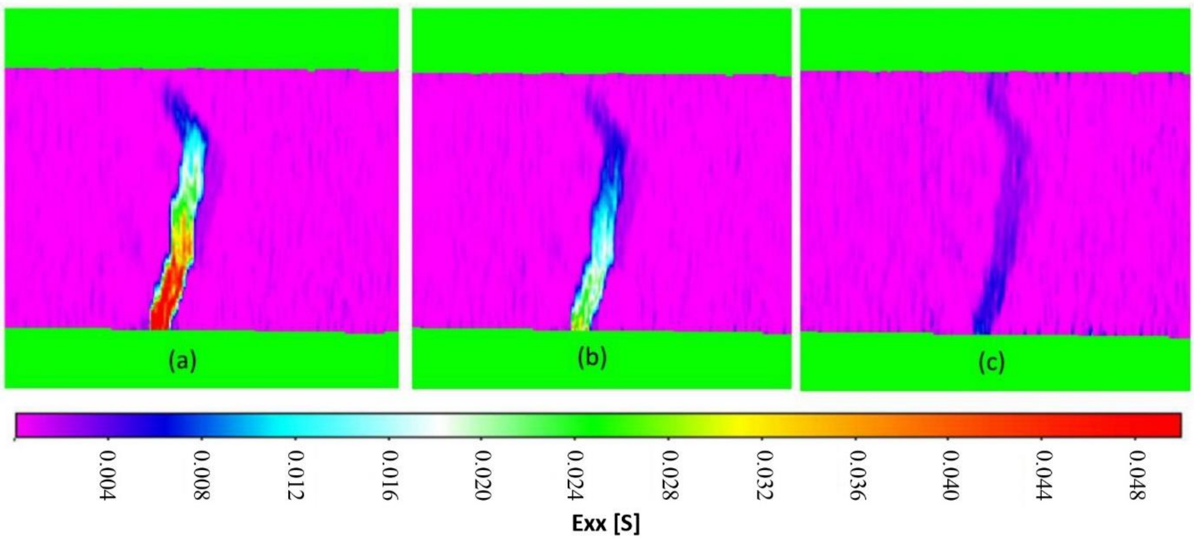


Figure 13. Digital image correlation (DIC) camera snapshots showing closure of unreinforced concrete crack using PET tendons. Axis indicates strain. (a) Loaded to 0.5 mm CMOD; (b) unloaded; (c) post-activation. [266]

4. Test methods to evaluate self-healing efficiency

The high number of varying test methods to measure the durability of healed mortar/concrete makes it difficult to compare and interpret data by different research groups. The standardization of methods for crack creation, curing procedures, and methods to evaluate self-healing efficiency, would contribute to obtaining reliable data and would allow for incorporating self-healing concepts into durability-based predictive models and design approaches.

A previous extensive literature review performed by Ferrara et al. [18] shows that there are multiple essential variables that influence laboratory experiments to assess self-healing concrete, such as the age of cracking, crack width, presence of water, the status of the crack (static vs active cracks), and repeatability. Beyond that, to measure durability, the performed tests depend on the main attack mechanism(s), which are related to the aggressive scenarios, and to the main transport mechanism of the aggressive substances. Often due to the combined action of aggressive agents or the variety of ingress mechanisms, evaluating an isolated parameter will not bring sufficient information regarding the durability performance. The determination of the effectiveness of self-healing inside the cracks is not straightforward, but confidence can be raised with suitable test methods.

In order to avoid duplication, the authors refer to the state-of-the-art report prepared by SARCOS COST Action CA15202 to learn more about the experimental characterization of the self-healing capacity of cement-based materials and its effects on material performance [18]. For the sake of completeness of the current paper, an overview is given (Table 3) of tests and methods for characterization of self-healing related to (1) crack closing quantification, (2) recovery of durability properties and (3) recovery of mechanical properties, mainly based on [18].

Table 3. An overview of methods (non-exhaustive) to measure the performance of healing agents with a focus on durability.

Test method	Purpose	Advantages	Influential factors related to execution and interpretation of the test	Limitations	Some references (non-exhaustive)
Crack closing evaluation					
Optical microscopy	- Assessing crack closing at the surface - Quantification of the change in crack width or area	- Non-destructive - Simple working principle and easily implementable - Monitoring crack closure in time and space (along the crack) possible	- Water on the surface can complicate the interpretation of the images	- No guarantee about the closure of the crack in deeper parts and the liquid tightness - No volumetric crack closing ratio can be obtained	[1,25,277,297,298,26,27,36,171,172,188,225,239]
Fluorescent microscopy and thin sections	- Assessing self-healing in depth	- Good visualization of crack healing over the complete depth	- Matrix fragility - Sample preparation	- Destructive testing - Upon extracting the sample, damage can be caused	[23,36,235]
Scanning electron microscope (SEM)	- Production of high-resolution images - Confirming the presence of healing products - Providing information about the chemical composition when combined with EDX (see below)	- Backscattered electrons (BSE): differences in the atomic number can give information on phases - Secondary electrons: topographic information	- Sample preparation (polished for BSE, unpolished for topographic imaging)	- Small samples and test area - Many images need to be studied for representative results	[23,24,300–309,26,310,168,172,188,204,235,297,299]
Energy-dispersive X-rays (EDX)	- Chemical identification of the self-healing products - Qualitative and quantitative analysis	- Self-healing products chemical identification	- Selected location: elemental mapping and line scans	- Small samples and test area	
X-ray computed microtomography (µCT)	- Defining damage, failure, porosity: volume of the crack and healing products - Defining the geometry of the crack - Determining the internal self-healing capabilities - Phase identification and density	- Non-destructive testing - Evidence of the damage - Quantification of the crack closure by self-healing processes	- Quality of data analysis	- Small sample sizes - Restricted to pastes and mortars	[168,184,188,297,311]
X-ray computed tomography (CT)		- Can be used at the concrete level		- Larger equipment	[310,312–314]
X-ray radiography	- Measurement of moisture uptake over time - Visualization of rebar corrosion in cracked and healed concrete	- Non-destructive testing - Images at high-spatial resolution	- Proper image overlap - Comparison between specimens in the dry and wet state	- Difficult to obtain a perfect overlap of the images before and after healing	[1,141,322,323,292,315–321]
Nuclear magnetic resonance (NMR)	- Qualitative and quantitative approach of water transport	- Non-destructive testing - Allowing a detailed study over time	- Regions filled with water can be confused with crack locations	- Small sample size (nm features) - Equipment not widely available	[324,325]

Durability tests					
Water permeability test	- Assessing the flow of water through healed cracks - Obtaining the water permeability coefficient - Assessing the recovery of water tightness	- Different pressures can be applied - Evaluation of the tightness - Measurement of the resistance against aggressive substance penetration	- Geometry, tortuosity and width of the cracks - Pressure of water - High water velocity can remove the healing products and causes inaccuracy in the test results	- Restricted to saturated elements	[25,36,236,239,257,285–291,155,292,293,172,184,194,212,213,224,225]
Gas permeability test	- Obtaining the gas permeability coefficient - Assessing the recovery of gas tightness		- Type of gas - Pressure of gas	- Set up with high demands for precise measurement control	[309,326,327]
Sorptivity test	- Rate of absorption (sorptivity) of water	- Much simpler, less time-consuming, and does not require a special device when compared to the permeability tests	- Sorptivity is not restricted to the crack but also via the matrix by capillary porosity - Tortuosity and crack geometry	- Restricted to dried elements - Interpretation of the results can be difficult in the case of partial crack healing, which may increase sorptivity in comparison to cracked and unhealed samples due to higher capillary forces	[48,181,329–333,194,201,221,252,290,310,322,328]
Chloride penetration	- Assessing the transport of chlorides and rebar corrosion risk - Determination of the Cl profile near the crack - Determining corrosion onset, Cl threshold	- Direct evaluation of the self-healing performance in chloride-containing environments	- Saturation degree of the concrete determines the transport mechanism - A choice should be made regarding the test method to evaluate Cl diffusion, absorption, migration	- Need for accelerated tests instead of realistic tests, which are time-consuming	[26,78,166,199,224,227,228,254,331]
Sulphate attack	- Evaluating the sulphate degradation	- Direct evaluation of the self-healing performance in sulphate-containing environments	- Sulphate concentration - Specimen size - pH - Temperature - Type of binder (amount of C_3A) - Presence of SCMs - Type of immersion (total, partial or wet-dry cycles)	- Need for accelerated tests due to slow evolution in case of realistic tests - Sensitive test methods must be used to estimate the damage when simulating real non-accelerated conditions	[86,113,231–233,245,334]
Carbonation test	- Assessing the transport of CO_2 and rebar corrosion risk - Determination of the neutralisation profile	- Direct evaluation regarding the carbonation resistance of self-healing cementitious materials	- Slow process in natural conditions - Risk of high %CO_2; artefacts by high %CO_2 - Relative humidity	- Need for accelerated tests to limit test duration - The concentration of CO_2 is suggested to be not more than 4%	[310]
Recovery of mechanical properties					
Ultrasonic wave propagation	- Detect concrete quality, internal defects, cracks and voids	- Simple working principle and easily implementable - Immediate results, portable, large penetrating power - Moderate cost	- Variability in the coupling between the sensor and the surface of the specimen leads to variability in results	- Qualitatively reliable - Moisture content, temperature changes, presence of reinforcement, and local crack bridging can affect results - Specimens that are rough, irregular in shape, very small, thin, or not homogeneous are difficult to inspect.	[24,167,192,252,261,335–339]
Resonance frequency	- Determining the rate and extent of self-healing	- Non-destructive testing - Quantification of the crack closure - Sensitive to small variations	- Further continued hydration can mask the self-healing efficiency - Fluctuations in the measurements due to the saturation changes	- Comparative analysis is defined by controlled features such as saturation and rate of damage	[311,340]
Mechanical recovery test	- Evaluation of recovery of mechanical properties	- Correlate the recovery of different mechanical properties (strength, stiffness, strain and deformation capacity) with crack closure. - Measures evolution of parameters useful for structural design	- Loading rate - Choice of characteristic points on the loading curve and choice of parameters used to calculate recovery indexes	- Comparative test needed on sound specimens - Measurement of the overall properties of the specimen - Need to distinguish between effects of concrete ageing and effects of healing	[18,24,309,329,338,341–346,31,171,177,179,186–188,237]
Acoustic Emission	- Continuous monitoring of crack initiation and propagation - Evaluation of healing - Monitoring release of water by SAPs	- Breakage of capsules, cracking, and healing can be monitored - Evaluation of the durability of concrete elements subjected to different loading types	- Positioning of sensors - Defects or early cracks blocking the wave travel path for later cracks	- Expensive - Defects already present are not detected	[36,236,291,310,336,339,347–349]

5. A pathway for the incorporation of self-healing performance into a life-cycle spanning durability-based design

The obstacles preventing the large-scale application of self-healing technologies in day-to-day construction practice are currently related, on the one hand, to the lack of standardized test methods to assess the self-healing performance and, on the other hand, to the complete absence, in structural design codes, of any pathway to fully recognize the effect of self-healing functionalities on material and structural performance.

In a broader sense, this is related to how durability is currently dealt within structural design codes. The options, as per fib Model Code for Service Life Design are: full probabilistic approach; semi-probabilistic approach (partial factor design); deemed-to-satisfy rules; and avoidance of deterioration [66].

1. The *full probabilistic approach* is based on probabilistic models for deterioration and material resistance. Statistical information on the variability (randomness) of the contributing parameters is required in this approach. These models should be validated to give realistic, representative, and reliable results. The method should also be based on appropriate test methods with statistical evaluation. Generally, the normal probabilistic distribution is used to account for the randomness of the parameters involved.
2. The *partial factor approach* uses statistically derived partial factors and is intended to be a practical, statistically reliable design tool. However, the partial factors are derived using the same models as the full probabilistic approach. Such data is obtained from a huge set of examples.
3. The *deemed-to-satisfy approach* is conceptually like current prescriptive durability specifications, based on a selection of design values (e.g. dimensioning, material and product selection, execution procedures).
4. The *avoidance of deterioration* approach requires the use of deterioration-resistant materials such as stainless steel, or concrete protection systems such as coatings, thus limiting or eliminating deterioration of the structure. Maintenance may still be required, such as renewal of coatings from time to time.

So far, national and international concrete standards provide requirements to achieve the desired design service life based on the “deemed to-satisfy” and the “avoidance of deterioration” approach, where durability-related exposure conditions are defined according to 17 exposure classes [60]. Nevertheless, fib Model Code 2010 [350] recognizes that “If more refined service life designs are to be undertaken by the use of deterioration modelling, this classification of the environmental load must be related to quantified parameters, e.g. chloride concentrations for marine structures. When publishing this Model Code, such quantified parameters were not available in any operational standard. The information must therefore be found by measurements on existing structures and in the literature”.

It is worth remarking that, even in the case of those deterioration mechanisms for which models with a relatively broad international acceptance exist, the codes consider the concrete as a passive provider of protection to the rebar rather than as an active player in the durability performance achievement. Verification of performance implies checking that the critical front of the deterioration mechanism does not reach, over the intended time, a depth equal to the reinforcement cover. No information is given on the time evolution of the material properties, and hence no information is obtainable on the residual level of “safety”, which characterizes the structure at the intended end of its service life. Moreover, the models proposed for the deterioration mechanisms explicitly considered in the codes, including carbonation-induced corrosion, chloride-induced corrosion and acid attack, do refer to sound un-cracked concrete, a state which hardly represents the real service conditions of reinforced concrete structures [55,60]. Though the verification of some durability-related limit states may include a crack width control, no information is provided on the evolution of such a width along the structure’s service life due to interaction with material ageing, deterioration or self-healing phenomena, which can accelerate or slow down the advance of the deterioration mechanism's critical front. This is also due to the fact that most of the currently available design codes have been originally formulated for normal strength (Portland cement) concrete and then further extended, with some limitations, to the broad category of high-performance concretes, which do actually have to include self-healing concrete.

A possible pathway to incorporate the benefits of self-healing into a durability-based structural design should, first of all, start from the quantification of a "scenario-dependent" healable crack width. In this respect, an attempt has been recently made to quantify the dependence of the healable crack width on a plurality of variables, including concrete composition, initial crack width, type and duration of the healing, through artificial neural networks [20]. Similarly, the effects of self-healing on the parameters governing the degradation mechanisms should be quantified, e.g. from an AI-based analysis of literature reviewed in this paper. The incorporation of the "self-healing affected" durability parameters and "self-healing delayed" degradation mechanisms into structural design algorithms could lead to the quantification of the benefits of using self-healing concrete in terms of extended design service life and reduced maintenance [351–353].

In addition, life cycle assessment (LCA) can properly help to support and to promote the deployment of self-healing concrete, assessing all the impacts from mining, production, and use until the possible end-of-life scenario [354,355]. Compared to LCA studies for traditional concrete, the scientific literature about LCA of self-healing concrete, is limited. As a matter of fact, previous LCA studies focused mostly on sustainable concrete options like concrete containing incinerator ashes, marble sludge, blast furnace slag, recycled aggregates or fly ash [50,356–359]. This was motivated by the fact that cement-based composites are often associated with huge negative environmental impacts: the production of Portland cement (PC) only, can lead to 2 billion tons/year of CO_2 emissions (approximately equal to 8-10% of global anthropogenic emissions) [360–363]. Therefore, considering the consistent annual production of concrete, higher than 10 billion tons globally [364–367], more sustainable solutions in this field can help to reduce negative environmental impacts related on a global, regional and local scale [368]. The cited studies demonstrated that both the cement content and the extended service life had a significant impact on the overall environmental performance. More specifically, 36%-43% and 36%-38% of carbon footprint and energy consumption, respectively were avoided in the study developed by Nath et al. [50] by replacing 40% of cement by fly ash and increasing the service life 1.6-1.75 times. Robayo-Salazar et al. [356] used a natural volcanic pozzolan and granulated blast furnace slag, reaching 45% carbon footprint reduction. Therefore, LCA can also prove that self-healing concrete can achieve significant benefits from an environmental point of view because of the resulting extended service life (SL) [225].

The LCA procedure is outlined by ISO 14040:2006, and 14044:2006 standards that identify four major steps: (*i*) definition of goal and scope (choosing a representative functional unit and considering an appropriate system boundary), *(ii)* inventory analysis, *(iii)* impact analysis and *(iv)* interpretation. A proper functional unit (FU) choice, as pointed out by Panesar et al. [49] and Van den Heede et al. [368], being the basis for estimating input and outputs, is mandatory to obtain reliable LCA results. Gursel et al. [369] highlighted the necessity to include most of the concrete performances like strength, durability and unit weight, while in Van den Heede et al. [368], extending the assessment by Damineli et al. [370], proposed a FU defined as "the total amount of binder per m^3 concrete necessary to deliver 1 MPa of strength and one year of service life" emphasising the link between LCA outputs and SL.

System boundaries, as summarized by Figure 14, could be: *(i)* cradle-to-gate (referring to the production process), *(ii)* cradle-to-grave (including, besides the "production phase" also the "use" and "end-of-life" stages) or, better, but not yet investigated for self-healing concrete *(iii)* cradle-to-cradle (which includes "production", "use", "end of life" and "recycling of the waste" phases) [371,372]. Wu et al. [373] warned to exclude "use" and "end of life" stages according to the ISO 24067 standard only if they affect LCA results by less than 1%. The importance of an appropriate system boundary is clarified further by De Schepper et al. [372], who demonstrated how, choosing an appropriate cradle-to-cradle system boundary for LCA of recyclable concrete, the recycling of concrete for clinker production can reduce the impacts by 8-15% in comparison to traditional concrete.

As mentioned above, service life and durability are crucial in LCA studies, but, unlike ordinary concrete, there is little literature on this subject. Caruso et al. [374] and di Summa et al. [375] investigated the effect of the extended durability with a resulting service life of at least 30% longer on LCA output, developing a cradle-to-grave analysis for innovative cementitious materials exposed to aggressive ambient conditions. Here the authors investigated, in the construction of a basin for geothermal water settling, a high-performance fibre reinforced cementitious composite with high content of slag (50% replacement by volume of cement) and 1.5% by volume of steel fibres (to obtain a strain hardening tensile behaviour) and autogenous healing stimulated by crystalline admixtures (CA). They estimated a reduction of 71% and 87% for abiotic depletion and acidification, respectively. Similarly, Van den Heede et al. [53] highlighted that the use of healing agents, together with PVA microfibres, allowed to save approximately 80% of global warming potential (GWP) if compared to Portland cement (PC) concrete due to the extended service life equal to 60 years (35 years longer than PC concrete).

Similarly, the recent results of di Summa et al. [353] are in line with what is mentioned above. More specifically, they demonstrated that in a timeframe of 100 years of service life, the concrete containing superabsorbent polymers (SAPs), favouring both internal curing and self-healing, performed better than the conventional solution. This was mainly due to the reduced frequency of the maintenance activities ensured by the innovative technology, and it allowed reductions up to 50% for some impact indicators. Moreover, the study outlined the importance of using accurate durability parameters to get more reliable LCA and life cycle cost (LCC) outputs. In this case, the value taken into account for the apparent chloride diffusion coefficient, D_{app}, had a major effect on the time of corrosion initiation of the reinforcement bars. This largely affected the forecast of the frequency of maintenance activities with the consequent environmental and cost implications.

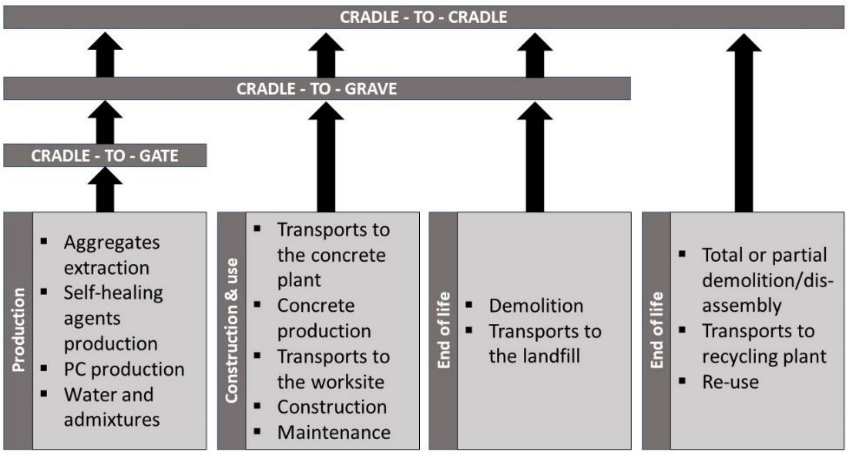


Figure 14. Possible LCA system boundaries for self-healing concrete.

A cradle-to-gate system boundary was used by Van den Heede et al. [52] for LCA of self-healing concrete containing different kinds of superabsorbent polymers. Here, the authors stressed two critical points, explaining why some studies are limited to the "gate" stage: *(i)* the limited knowledge regarding the service life when exposed to a specific environmental condition and *(ii)* the lack of data regarding the life cycle inventory (LCI). Because of the absence of data related to the healing agent (due to confidentiality issues of the manufacturer), the authors combined data from the *Ecoinvent* database with information reported in the literature. Furthermore, being aware that the development of new materials in the laboratory (as for most of the healing agents currently used) is not an energy-efficient process, they adjusted the LCI input by adopting the proposal by Piccinno et al. [376] to go from the laboratory to the industrial scale. The same challenges were faced by di Summa et al. [353], who also adopted a previously mentioned cradle-to-gate system boundary in the study. This was mainly due to the lack of information regarding the disposal scenario of the innovative composite. Moreover, even in this case, most of the LCIs have been redrafted according to existing scientific literature or to the environmental product declarations (EPDs).

Additionally, Hafez et al. [377] underlined that the uncertainties of LCA are also due to the fact that there are no standards to get LCI data. Therefore, they recommend following the suggestions by Anand et al. [378] to use either primary sources (like environmental product declarations or laboratory results), when possible, or secondary data from accredited environmental databases like Ecoinvent, Gabi and ELC. In addition to accurate data input, a representative impact analysis is essential. Jolliet et al. [379] defined the impact analysis as the touch point between LCI results and the corresponding environmental impacts that can be divided into two main impact categories:

(i) the first one, defined as problem-oriented, regroups specific environmental problems into specific midpoint categories, like the impact on climate change expressed in kilograms of CO_2 equivalents (e.g. CML 2002).

(ii) the second one, defined as damage oriented, tries to estimate the actual environmental damage, sometimes with high uncertainty (e.g. Ecoindicator 99).

The damage-oriented impact category promotes an easier to understand interpretation of LCA, but, as described by Benetto et al. [380], the problem-related approach is more reliable.

All of the available literature for LCA studies for self-healing concrete uses a problem-related approach assessing up to 10 indicators in total (as exemplified in Figure 15): global warming (GWP); acidification (AP); eutrophication (EP); ozone depletion (ODP); photochemical oxidation (POCP); abiotic depletion potential (ADP); human toxicity potential (HTP); freshwater aquatic ecotoxicity potential (FAETP); marine aquatic ecotoxicity (MAETP) and terrestrial ecotoxicity potential (TETP). All the studies estimate promising results for self-healing concrete from the environmental point of view. The investigation by Rigamonti et al. [54], which used a cradle-to-gate system boundary, cast a light on the better performance in terms of environmental impacts of self-healing concrete containing a crystalline admixture as an autogenous healing stimulator. In fact, impact indicators like AP, ODP, EP and POCP were reduced. This was also the case in the study by Van Belleghem et al. [225], where the same indicators were reduced by more than 50% because of the extended durability of concrete healed by encapsulated polyurethane.

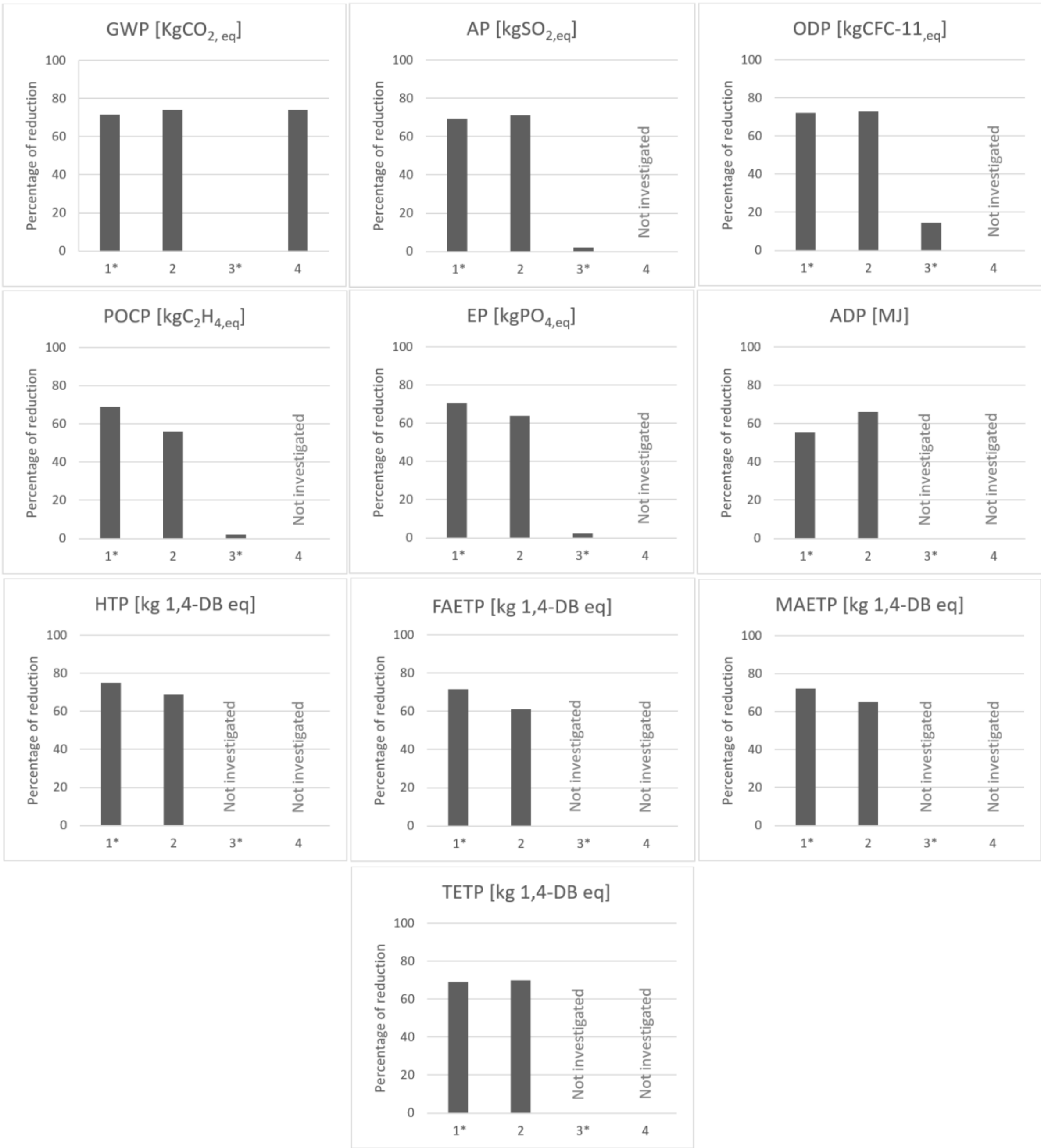


Figure 15. Comparison of the reduction of impacts among the available literature about LCA of self-healing concrete. (1) concerns concrete composites with 1 mass% of SAP and 2 vol% of PP microfiber, taking into account a FU with 100 years of service life [52]*; (2) similarly, but for self-healing concrete with polyurethane* [225]*; (3) concerns concrete with crystalline admixture using a cradle to gate system boundary* [54]*; (4) concerns a concrete composite with 1 mass% of SAP and 2 vol% of PVA microfiber assuming 60 years of service life* [53]*. * data are obtained from "Figure 2" and "Figure 4" of the corresponding papers, respectively, as the exact value is not reported.*

Due to the uncertainties associated with data quality, some studies highlighted the necessity to process further the results obtained by LCA analysis using a stochastic modelling approach, known as Monte Carlo simulation, to estimate the uncertainty and communicate the results in a probabilistic way [50]. Even in Van den Heede et al. [53], considering the uncertainties related to maintenance and durability, the authors assumed a standard triangular error distribution that confirmed the sustainability of self-healing concrete with SAP (1 mass%/kg cement) and PVA microfibre (2 vol%).

Finally, regarding the Life Cycle Cost Assessment for self-healing concrete, only very few studies assessed the economic advantages of self-healing concrete compared to a conventional solution. In a work focused on the use of shape memory polymers to favour the autogenous healing, Teall already addressed the potential costs reduction within a service life of 120 years due to the reduced maintenance activities [381]. In line with this, two other recent studies have highlighted the influence of the repairing activities throughout the anticipated service life T, the cost of which for both the conventional and the self-healing solution, complying with ISO 16627:2015, has to be "actualized" through the annual real discount rate *r* according to the following equation [353,382].

$$(T)=1/(1+r)^{T}$$

This highlights the need to further analyse these innovative materials from an economic point of view, the reason why Caruso et al. [383], Gursel et al. [369] and Colangelo et al. [358] supported the idea of assessing thoroughly social and economic performances to achieve a more holistic sustainable approach combining the environmental implications with the ones that these composites can have in the society.

Conclusions

Self-healing is a promising solution to make concrete structures more durable. The aim is that cracks heal without intervention, increasing the service life and decreasing resource use, contributing to reducing environmental impacts and overall maintenance costs. Many self-healing mechanisms have been investigated and are available, but in real conditions, the most suitable one should be selected for the specific situation. Under ideal conditions, the healing efficiency of different self-healing systems has been widely studied. Actually, the "ideal condition" depends on the self-healing mechanism; however, it mainly refers to unstressed specimens and young specimens subjected to a single cracking/healing event, with the availability of water and at room temperature. The results obtained in this way may not be transferrable to real structural service scenarios, imposing limitations on their widespread use in current routine construction practice. Bridging the gap to move from ideal laboratory conditions to realistic field exposure should now be the focus.

Durability indicators, both general and specifically related to a specific degradation mechanism, should be determined to ensure the efficiency of smart concrete developed with self-healing ability. This is needed to incorporate self-healing into durability-based structural design approaches consistently. In this respect, it is of the utmost importance to focus on standardising tests capable of measuring the efficiency of self-healing mechanisms and quantifying their effects on the material mechanical and durability performance indicators. Artificial Intelligence may support this purpose, as currently done in several other fields of civil engineering, to "extract" design-oriented knowledge from widely flourishing literature, the information from which must be screened and guided to the goal.

It can be surely recognized that the incorporation of self-healing into a truly performance-based design is likely to bring economic benefits in terms, e.g., of reduction or optimization of concrete covers or shrinkage reinforcement thanks to crack healing and reduced values of permeability to aggressive agents. Furthermore, the actual benefits of self-healing, mostly related to the extension of maintenance-free lifespan and the reduced frequency of maintenance over the intended service life, have to be assessed in the framework of a life-cycle-based approach. Due to the large variability in the nature and costs of the aforementioned maintenance operations, very few studies have assessed the economic benefits of using self-healing concrete with reference to suitably defined "functional units" (structures or structural elements). To this aim, a closer collaboration between all the players of the (concrete) construction industry has to be sought, from researchers to major stakeholders, for a precise setting of the scope and goals. Efforts should range from standardization of test methods to formulation and validation of design methodologies and life-cycle assessment criteria to promote the widespread use of self-healing concrete in practice.

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The authors report there are no competing interests to declare.

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