



Laval (Greater Montreal)

June 12 - 15, 2019

SELF-HEALING CONCRETE: A REVIEW OF RECENT RESEARCH DEVELOPMENTS AND EXISTING RESEARCH GAPS

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Abstract: Self-healing materials are defined as the materials that are able to partially or completely restore their original functionality after they have been damaged. In cementitious materials, such as concrete, this concept is referred to the capability of material to seal the cracks without any manual interventions after damage, accompanied by regaining of the mechanical properties, which results in a more durable and sustainable structure. In addition to the available literature which fully describes different methods of applying self-healing into cement-based materials and evaluates their efficiency, new approaches and novel techniques have been proposed for this purpose. This paper presents a brief review of both autogenous and autonomous mechanisms of self-healing, with an emphasis on the recent research advancements. Since the major concern regarding the application of autonomous self-healing in concrete structures is the additional cost caused by the healing agents, the improvement of autogenous self-healing which is an inherent performance of cementitious materials by providing a favorable condition for the process (e.g. high-performance fiber reinforced cementitious composites) may pave the way towards construction industry. However, there are still a few aspects of self-healing concrete missing in the literature that inhibits this technology from being utilized in the construction. Therefore, a comprehensive section is proposed in this paper discussing the major gaps and outlooks in the field of self-healing concrete.

1 INTRODUCTION

Concrete is a frequently used construction material worldwide due to its relatively low cost and design flexibility. Despite its proven advantages, concrete often exhibits cracks which appear due to excessive tensile stresses or environmental conditions. These cracks are allowed to happen and are not considered as a failure in reinforced concrete (RC) members, as long as their width does not exceed a specified criterion. However, via these cracks, aggressive substances enter into concrete causing degradation and reinforcement corrosion. As a result of these phenomena, durability and mechanical performance of the concrete structure is compromised and its service life will be reduced. Therefore, it is vivid that the monitoring, maintenance, and repair of concrete cracks is of great importance.

However, the continuous inspection of cracks and repairing them is difficult, especially when it comes to cracks in infrastructures such as bridges, highways or tunnels. In spite of the fact that cracks in infrastructures are not easily accessible and require a considerable amount of labor and budget, many of these infrastructures are in continuous service which makes repairing even more difficult. It was also reported that in Europe, 50% of the annual construction budget is spent on repair works to prolong the service life of the structures (Cailleux and Pollet 2009). Apart from the high costs demanded for repair, most of the conventional repairs can only last for ten to fifteen years (Van Breugel 2007). Under such circumstances, the most effective method for repairing the cracks is to provide an automatic healing

mechanism that triggers upon necessity and regains the functionality of the concrete structure without human intervention.

Self-healing mechanism is defined as “any process by the material itself involving the recovery and hence improvement of a performance after an earlier action that had reduced the performance of the material” (De Rooij, Van Tittelboom et al. 2013). Different mechanisms of self-healing have been introduced during the years, however the history of self-healing phenomena can be tracked back to ancient structures and buildings, where cracks being filled with white crystalline materials were observed (Nijland, Larbi et al. 2007, Ghosh 2009). This intrinsic property of concrete that fills the cracks with hydrated pozzolanic binders or crystalline materials in the proper condition is called “autogenous self-healing”. By realizing the whole concept that concrete is capable of sealing and healing the cracks without external activation, the researchers were persuaded to develop “Autonomous self-healing” by designing and adding engineered materials into concrete to act upon cracking. They aimed at tailoring novel mechanisms that expected to be more efficient than autogenous self-healing. However, the initial cost for these techniques used in this category rises a significant concern for its industrial application. To overcome the mentioned impediments, researchers came up with a self-healing mechanism called "Improved autogenous" that follows the basics of autonomous self-healing, i. e. engineered conditions, yet the whole function and healing products are identical to autogenous self-healing. Figure 1 which is essentially based on Venn Diagram of self-healing/repairing of cracking reported by the technical committee TC-075B from the Japan Concrete Institute (JCI) (Igarashi, Kunieda et al. 2009), explains the relationship between different major self-healing mechanisms.

The scope of this paper is to present a brief review of all major mechanisms for self-healing in cementitious materials, also express the most recent works on every mechanism and highlight their flaws and strengths. Based on the existing research gaps, outlooks for further investigation on self-healing in engineering practice are proposed.

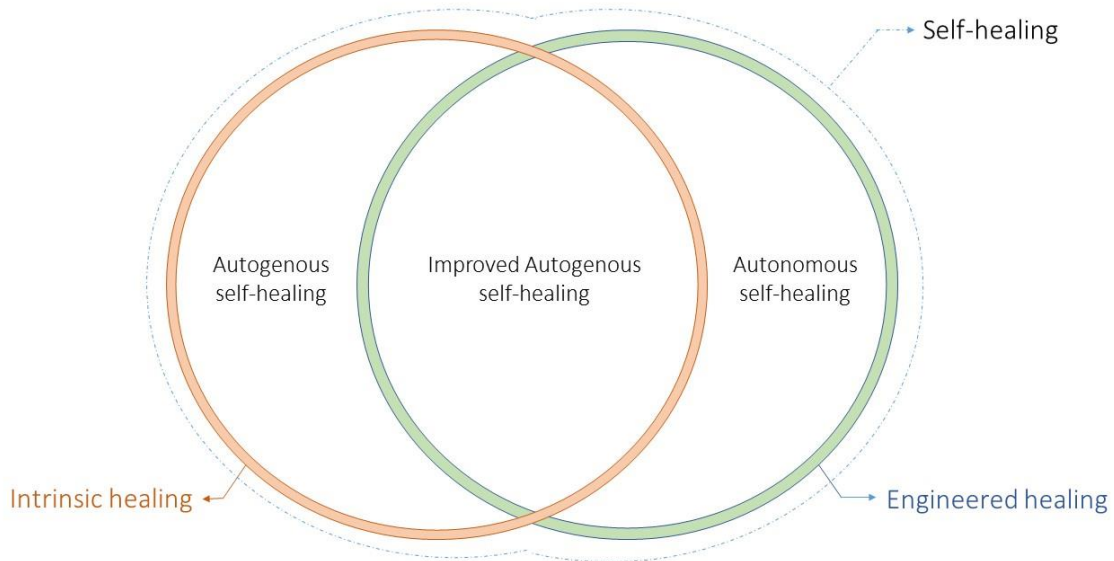


Figure 1: Schematic diagram illustrating self-healing mechanisms

2 AUTONOMOUS SELF-HEALING

RILEM technical committee 221-SHC (De Rooij, Van Tittelboom et al. 2013) defines autonomous self-healing as the process of healing with the material components that are specifically in concrete for this purpose (engineered additions). Utilization of bacteria that results in the precipitation of calcium carbonate (CaCO_3) when contacted with water (De Muynck, De Belie et al. 2010, Jonkers, Thijssen et al. 2010, Van Tittelboom, De Belie et al. 2010, Wiktor and Jonkers 2011), mineral admixtures that are capable of forming crystalline or expansive productions in response to a crack (Ahn and Kishi 2010, Jaroenratanapirom and Sahamitmongkol 2011, Qureshi, Kanellopoulos et al. 2016), and encapsulations of chemical healing agents (Van Tittelboom, De Belie et al. 2011, Yang, Hollar et al. 2011, Hilloulin, Van Tittelboom et al. 2015) are the main methods of autonomous self-healing that have received the bulk of attention from researchers in the past decades. Despite the promising results that have been reported, the high initial cost of some of these techniques keeps them off the table when it comes to industrial application. Cultivation of bacteria or tailoring encapsulation techniques with materials that should withstand the harsh and alkaline environment of concrete, may sometimes become more expensive than expected (De Muynck, De Belie et al. 2010, Dong, Wang et al. 2015, Silva, Boon et al. 2015). Still, novel approaches were carried out by researchers in this field that indicates the potential that lies in this mechanism.

Among all autonomous methods, the pace of development in bacteria-based healing has been striking. Now researchers have identified types of bacteria that not only show reliable results in self-healing of cracks; they can also enhance the strength properties of concrete (Nain, Surabhi et al. 2019). Moreover, Nanomaterials have been introduced in the last few years as a suitable complementary mechanism for self-healing in concrete (Lee, Yoon et al. 2016, Huseien, Shah et al. 2019). Faster hydration rate for cement and other pozzolanic binders, reduction of porosity, plus improved interfacial bonding between hardened cement paste and other components has been reported to be the main effects of nanomaterials on concrete, which also have positive effects on self-healing as well (Zhang and Islam 2012). Nevertheless, other applications such as inner water providing by nanocalys or increased formulation of C-S-H by nanosilica were found to be also effective on self-healing process (Qian, Zhou et al. 2010, Sikora, Abd Elrahman et al. 2018).

3 AUTOGENOUS SELF-HEALING

Autogenous self-healing can be considered as the pioneer of self-healing mechanisms since this phenomenon was first observed about 200 years ago in water retaining structures and pipes by the French Academy of Science (Lauer 1956). Further hydration of unhydrated cement, calcite precipitation, and swelling of cement paste are of the intrinsic characteristics of ordinary concrete that all together could perform as a barrier against ingress of harmful substances, or might even regain the mechanical properties of concrete in a favorable environment (Figure. 2). However, the mentioned characteristics bring up limitations for this mechanism as well. It is reported that almost 80% of the hydration process in concrete is completed by the first 28 days. Besides, the formation of calcite which is the primary cause of autogenous self-healing at later ages (Neville 2002) is extremely dependant on the surrounding environment (e. g. Presence of water or moisture is mandatory; also sufficient carbonates or bicarbonates should exist to react with Ca^{2+} ions). The shape of the induced crack is also influential on the results (Ter Heide 2005). All these uncontrollable effective parameters may explain the reason for the wide range of healed crack width (5 μm to 300 μm) reported by researchers due to autogenous self-healing (Clear 1985, Jacobsen and Sellevold 1996, Reinhardt and Jooss 2003).

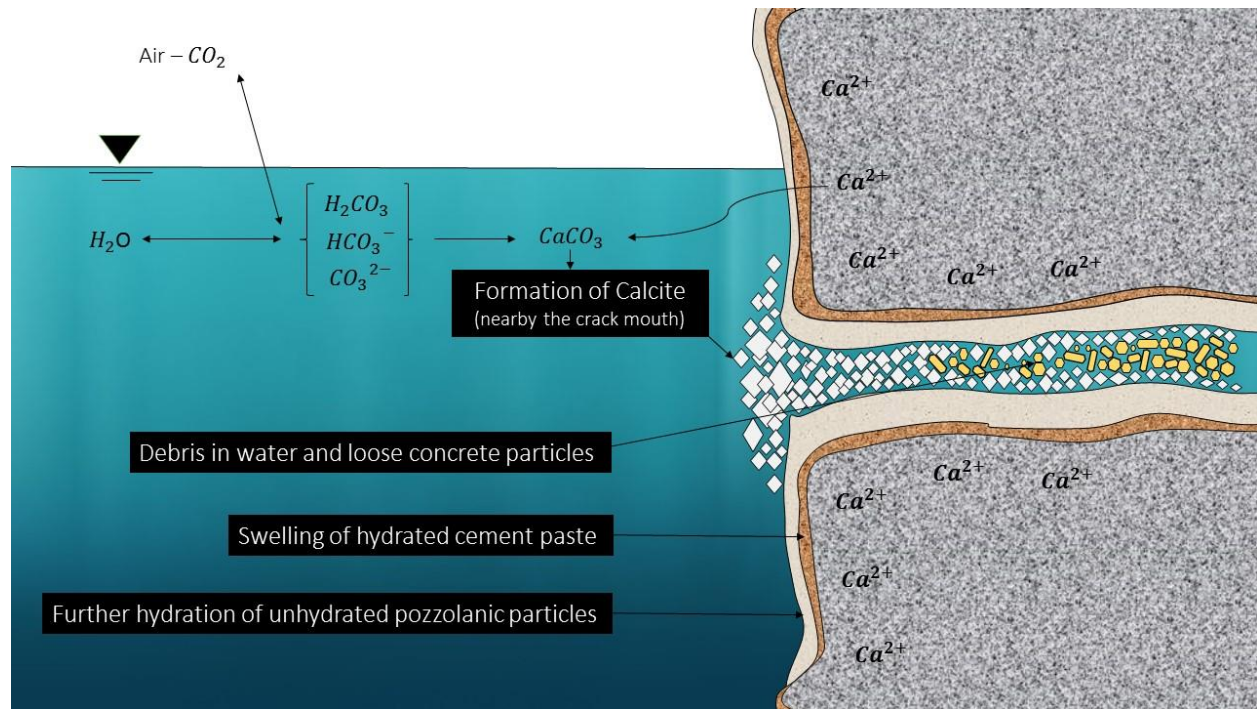


Figure 2: Main mechanisms of autogenous self-healing in concrete

4 IMPROVED AUTOGENOUS SELF-HEALING

Since the entire process of autogenous healing is quite recognized to the researchers, favorable conditions have been engineered to improve the limitations discussed in the previous section. As mentioned, the crack geometry is of utmost importance while assessing the autogenous self-healing efficiency; thus, tailoring fibres in concrete such that results in microcracking behavior (i. e. restricted crack widths ($\sim < 100\mu m$) and exhibiting a semi-uniform distribution of microcracks instead of a major crack) will significantly enhance the intrinsic self-healing of concrete (Homma, Mihashi et al. 2009, Cuenca and Ferrara 2017). Moreover, by replacing the cement content with other pozzolanic binders (e. g. fly ash and Ground Granulated Blast Furnace Slag (GGBFS)) that demonstrated to have slower hydration rates, autogenous healing based on further hydration will be promoted even in later ages (Sahmaran, Yildirim et al. 2013). Also, compatible supplementary materials that result in crystallization (CA) and Super Absorbent Polymers that provide water into the cracks (SAP) have been reported to be effective in terms of autogenous healing (Ferrara, Krelani et al. 2016, Park and Choi 2018, Tenório Filho, Snoeck et al. 2018).

Engineered Cementitious Composite (ECC) is a special category of High-Performance Fiber Reinforced Cementitious Composites (HPFRCC), introduced by Victor Li and his co-workers at University of Michigan (Li 1998). The main characteristic of ECC that distinguishes it from other fiber reinforced cementitious composites is its strain-hardening behavior that results in desirable inelastic deformations and multiple crack bridging with control property under tensile stresses (Figure 3). A typical ECC mixture consist of cement, flay ash/slag, sand, superplasticizer, Polyvinyl Alcohol (PVA) fibers and water. The fibers and ingredients in ECC are tailored based on micromechanics material design theory, which results in a synergetic interaction between fibers, cementitious matrix and the interface (Li 1998, Yıldırım, Keskin et al. 2015). Considering the well-controlled crack width behavior and the fact that fly ash/slag do exist in the mixture of ECC, this composite material has been preferred by many researchers for its promising results on improved autogenous self-healing.

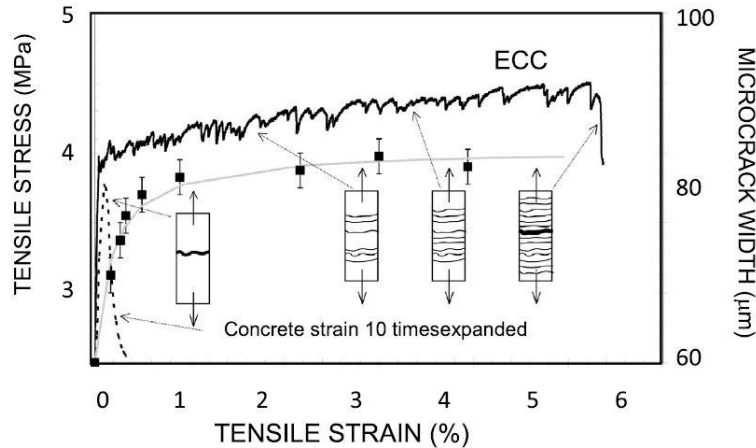


Figure 3: Stress-strain curve for ECC with crack width development and crack pattern (Şahmaran and Li 2009)

Several investigations have been carried out toward determining the optimum mixture and conditioning for ECC. The main variables considered for this purpose included fiber's material type and fraction (Homma, Mihashi et al. 2009, Snoeck and De Belie 2012, Snoeck and De Belie 2015), binder replacement (Özbay, Sahmaran et al. 2013, Sahmaran, Yildirim et al. 2013), the age of preloading and exposure condition (Li and Li 2011). A recent approach in this regard was replacing the cement by environmental friendly Reactive Magnesia-based Cement (RMC) in strain hardening composites, which proved that crack sealing and significant mechanical recovery could be achieved through proper environmental conditioning (Qiu, Ruan et al. 2019).

5 RESEARCH GAPS AND OUTLOOKS

5.1 Repeatability

All the available self-healing mechanisms demonstrated to be "feasible" to different extents; however, it is necessary for every individual mechanism in a sustainable infrastructure to be "robust" as well. To assure a healing mechanism is robust, it needs to meet at least six robustness criteria including shelf life, pervasiveness, quality, reliability, versatility, and repeatability (Li and Herbert 2012). Despite highlighting the advantages and weaknesses of every approach, the robustness criteria could be considered as a filter for these approaches, which identifies the practical methods for self-healing in infrastructure.

Among all six, the repeatability criterion raises a significant challenge into the healing mechanisms due to the inevitable repeating nature of different external loads over the lifetime of the infrastructure, causing repetitious cracking. Therefore, a robust healing method is required to be able to function not just once, but to trigger upon multiple cracking over the lifetime of the infrastructure. Micro and macro encapsulating techniques employed for self-healing based on bacteria and adhesive agents have shown promising results regarding their healing functionality, yet they are less likely to be repeatable. The repeatability index for these methods highly depends on their proportion in the cement matrix. However, high ratios of healing agents may result in strength degradation (Khaliq and Ehsan 2016), plus it increases the initial cost of producing a robust self-healing concrete which is still a substantial concern for the commercial use of these type of materials.

With the development of Engineered Cementitious Composites in the last few years, the repeatability of improved autogenous self-healing has brought the attention to the researchers. M. Li (Li and Fan 2016) confirmed that for a self-healing mechanism to be repeatable, a set of physical, chemical and environmental conditions must be satisfied. Fibers that bridge the cracks to control the damage and provide nucleation sites, plus high pozzolanic ingredients and unhydrated cement that promote the formation of C-S-H- are

suggested to be most effective. A thorough study carried out by Sahmaran (Sahmaran, Yildirim et al. 2015) on the repeatability of improved autogenous self-healing in ECC proved that under certain condition (i.e., the presence of water), the healing method demonstrates acceptable results even after nine cycles of loading and unloading. V. Li (Li and Herbert 2012) also claimed that Improved autogenous healing in ECC is repeatable in the natural environment, but highly dependent on the amount of damage and weather conditions. To overcome the water-supplying problem of autogenous self-healing in repeated loadings, D. Snoeck (Snoeck and De Belie 2015) implemented SAP (Super Absorbent Polymer) in strain-hardening cementitious composites. He compared the results for two consecutive loading and unloading cycles and found out that SAP can play a significant role in the regain of mechanical properties of the damaged concrete under repeated loads.

Nevertheless, it is difficult to determine a single load pattern in lab that simulates the exact condition of the external repeating loads. Since almost all of the research conducted on assessing the repeatability of self-healing mechanisms employed a simple series of loading and unloading, it is possible that with a more precise loading approach, different results would be achieved not only for the repeatability index, but also the whole robustness criteria. Different magnitudes, frequencies and directions of loading should be tested for this purpose. Unfortunately, the lack of field studies in this case expands the existing research gap.

5.2 Self-healing in reinforced concrete

Majority of the experiments carried out toward demonstrating the performance of self-healing methods were based on small-scale unreinforced specimens. However, since most of the concrete structures are reinforced, it is essential to consider the possible difference in self-healing process and outcomes while the steel is present. In one of the few researches in this regard, Keskin et al. confirmed the effectiveness of intrinsic self-healing of ECC in large scale reinforced beams (Keskin, Keskin et al. 2016). It is reasonable to believe that the difference is negligible, yet two main concerns with respect to reinforced concrete is the physical and chemical reactions due to corrosion that can manipulate the self-healing process and also the plastic deformation in steel which causes permanent deformations in concrete. To cover the research gaps mentioned, a thorough research should be conducted to assess the effects of corrosion process on each of the healing methods and also evaluate the effectiveness of employed methods in case of sustained cracks.

5.3 Effects of surrounding environment

Concrete members serve under different environmental exposure conditions based on their role in the structure. Through extensive investigations carried out in the last decades, the fact that the effect of the surrounding environment is inevitable on almost all of the self-healing mechanisms has been well established (Huang, Ye et al. 2016, Suleiman and Nehdi 2018). Therefore, adoption of a unique and effective self-healing method for a concrete structure highly depends on the exposure condition that the structure is expected to face throughout its service life. For example, while the presence of water is necessary for some of the self-healing mechanisms such as autogenous and improved autogenous healing and self-healing based on bacteria, it could have negative impacts on self-healing based on adhesive agents, due to the fact that water may inhibit the release of adhesive agents into cracks.

Usually, the curing conditions employed in the lab for assessing the effectiveness of a self-healing method are either based on their potential positive effects on the self-healing process or they are representing the anticipated field of application of the structure. Exposure to local climate, submerging in water, applying freeze/thaw and wet/dry cycles, controlled temperature and relative humidity are the most common curing conditions investigated by the researchers (Qian, Zhou et al. 2010, Zhu, Yang et al. 2012, Suleiman and Nehdi 2018). By collecting more in-depth knowledge of the surrounding environment and its effects on self-healing, the favorable self-healing mechanism can be adopted, plus in some cases, the surrounding environment can be tailored or promoted in a way that enhances the self-healing function. The studies carried out by Yildirim (Yıldırım, Khiavi et al. 2018) proved that increasing the concentration of dissolved CO_3^{2-} ions in the surrounding water of ECC specimens promotes the precipitation of calcite which is the

main source of autogenous healing at later ages, even for crack widths more than 200 μm (Fig. 4). The mentioned curing condition can also be considered as a nutrient-rich environment for bacteria that produces the same healing product (Huang, Ye et al. 2016).

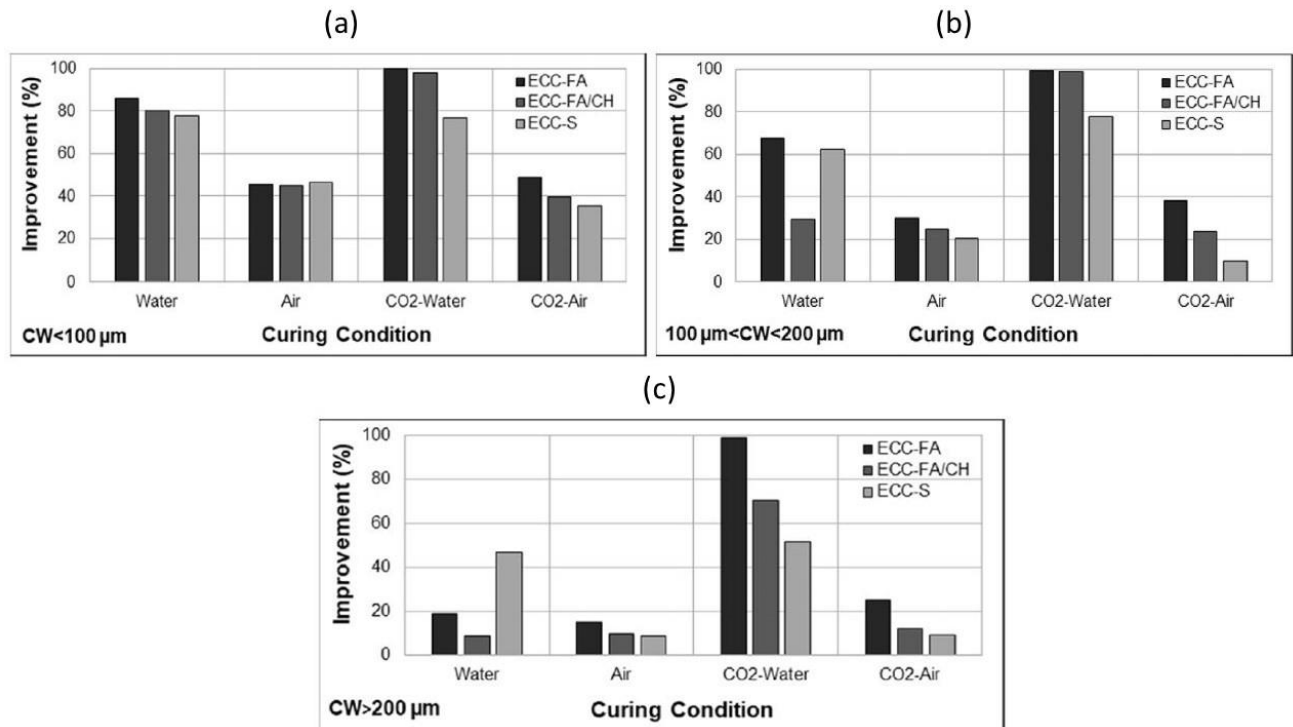


Figure 4: Self-healing improvement in ECC specimens under multiple curing conditions and different crack widths; (a) $CW < 100 \mu\text{m}$, (b) $100 \mu\text{m} < CW < 200 \mu\text{m}$ and, (c) $CW > 200 \mu\text{m}$ (Yıldırım, Khiavi et al. 2018)

However, the effects of different type of soils as the surrounding environment of a concrete member have been missing in the literature. Concrete pipes, tunnels, and also foundations of concrete and steel structures are of the main structural members that have soil as their surrounding environment through their lifetime. Since soil is full of chemical substances, and these substances may transfer into cracks by dissolving in the groundwater or water due to rain, there is a high possibility that by examining soil as the surrounding environment, different types of soil demonstrate different results for self-healing of cracks. To cover the research gap here, a thorough study requires to be carried out to identify the proper types of soil that exhibit the highest improvements in the self-healing results of cracks.

6 CONCLUSION

A review of recent research developments and existing research gaps has been presented in this paper. With considering both cost and efficiency, improved autogenous healing can be claimed to be the optimum mechanism for now. Autonomous healing is efficient in crack healing (300 μm to 1 mm), but the initial cost is still a matter. Autogenous healing is a free method but requires time and is highly dependent on the surrounding environment. On the other hand, the initial cost for Improved-autogenous healing is not considerable compared to autonomous healing, plus it performs in a reasonable time and heals crack up to 100-150 μm (Good results for ECC). However, recent achievements in bacteria based healing and nanomaterial treatment for self-healing in concrete may be able to pave the way towards industrial application, since they are offering a full package; a more sustainable, environment-friendly self-healing concrete with higher performance.

As mentioned, robustness is a crucial factor for a self-healing mechanism in sustainable infrastructure. The six robustness criteria reviewed in this paper (specially repeatability), have to be considered while employing a self-healing mechanism for sustainable infrastructure. Moreover, since researchers tend to develop engineered conditions into concrete to improve its self-healing behavior, studying the surrounding environment for possible engineered condition that boosts the intended healing mechanism is of high importance. Besides the research gaps mentioned in this paper, field study reports regarding self-healing mechanisms and a standard/code that defines the benchmarks for the quality of an acceptable self-healing mechanism is still missing from the literature. In addition, to establish a standard/code for self-healing, it is necessary to study the most common assessment approaches as well.

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