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A Decade of Research on Self-Healing Concrete

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Abstract

The main findings of a decade of research on the design and development of the first self-healing concrete are summarized in this chapter. The autonomous healing concept is introduced, and plethora of design campaigns is enlisted. Healing agent encapsulation and agent tubes vascular networks are reported as the most efficient healing configurations for laboratory-scale and real-size applications, respectively. Crack formation, closure after healing and further damage are phenomena tracked by using advanced experimental monitoring methods and their performance is critically revised. The effect of self-healing technology on concrete mechanical response, durability and long-term response to damage are critically discussed. The study contributes to the open discussion in the scientific research community regarding self-healing concrete upscaling feasibility and finally it aims to contribute as a base for the future studies dealing with concrete design optimization.

Keywords: concrete, cracking, repair, autonomous self-healing, damage monitoring

1. Introduction

1.1. A millennium of experience on concrete healing

The invention of construction materials equally strong and efficient to natural ones (i.e. stone, wood and fibers) is an inherent need of humans. Archeological findings prove that the concept of 'concrete' composite material is dating back to Roman Empire period. A variety of pozzolana compositions were established and numerous still standing today monuments are made of it, indicatively the Colosseum in Rome and the Mediterranean Caesarea harbor. The latter Roman concrete infrastructure is built undersea and its revolutionary composition is recently decoded: mineral aluminum tobermorite is exposed to sea water during the service life leading to progressive and post-curing lime crystallization [1]. The composition is reported as

the first historical evidence of concrete design that concerns self-healing mechanisms in an attempt to enhance its durability.

Durability issues arise in concrete when under early age shrinkage or service loads, micro-cracks form and propagate on the cementitious media. Due to material quasi-brittle nature and heterogeneous multi-scale composition, micro-cracks build dense networking, making macro-crack formation inevitable. This complex material response to damage is acquainted since antiquity, therefore smart lime crystallization mechanisms as in Roman concrete were introduced.

Extended literature exists on the autogenous repair mechanisms intrinsically built in concrete dependent on the material composition. The micro-organisms calcite precipitation and the continuous hydration of un-hydrated cement grains are the most promising crack repair mechanisms [2, 3]. Nowadays, the intrinsic autogenous hydration ability of cement is enhanced by embedding superabsorbent polymers at casting [4] and use of high volume fractions of micro-fibers (i.e. engineered cementitious composite [5]). Eventually, autogenous healing delays cracking, but it cannot effectively eliminate the degradation in the presence of macro-cracks. Therefore, the treatment of cracks with size of several hundred micrometers appears the crucial repair mechanisms and plays a key role on the material sustainable design.

1.2. The pioneering autonomous healing concept

Reaching the end of the previous century, technological progress has established easy and cost-effective methods to repair open macro-cracks on concrete. European Standard EN1504 provides guidelines on repair based on adhesive polyurethane or other epoxy agent injection into cracks. Today one finds plethora of conventional repair kits in the market that promise sealing of cracks ranging from capillary to few millimeters opening size. In real cases, the effective damage repair is limited since the agent cannot penetrate and effectively polymerize into narrow crack paths. Furthermore, only the cracks that reach the concrete surface, are visually detected and are easily accessible, can be treated using external agent injection. As a result, repair kits provide partial sealing and limited short-term repair, being therefore not a sustainable healing solution.

The pioneering research at the University of Illinois introduced the idea of autonomous healing design both on concrete [6, 7] and polymer composites (White protocol [8]). Dry et al. designed brittle fibrous tubes that carried sealant agent and embedded them into large concrete plates. The cracks developed due to surface shrinkage, propagated through the tubes and broke them releasing the sealant into the crack void. The cracks were filled and repair joints were formed at the concrete surface proving successful sealing. The healing prototype design was poorly investigated from the following years until 2009, when a boost in research studies on autonomous healing appears (see statistical graph in **Figure 1**).

1.3. Boost on research interest: design and composition optimization

Stepping on pioneering work of Dry, Van Tittelboom et al. designed laboratory-scale concrete beams carrying methyl methacrylate and polyurethane agent embedded into short (typical

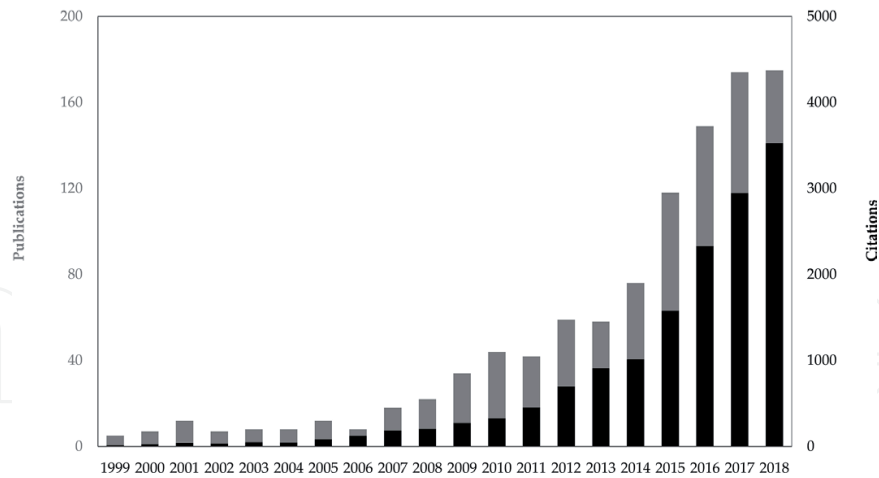


Figure 1. Total publications (right vertical axis) and summary of times cited (left vertical axis) per year in the topic of 'healing concrete'. Source: Web of Science database.

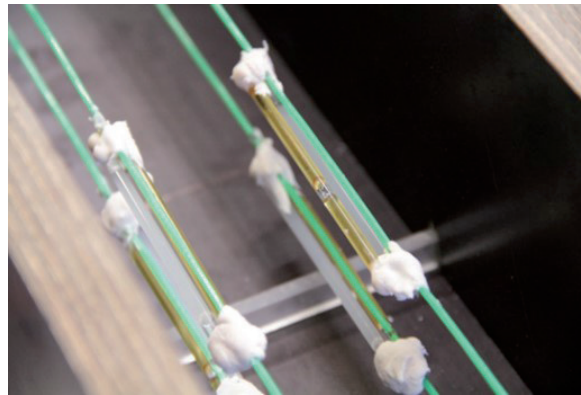


Figure 2. Four pairs of borosilicate glass capsules carrying the healing agent (polyurethane in yellow) and its actuator (white component) are attached using thin plastic wires to the concrete mold. Picture taken before concrete casting.

length at 50 mm), thin (diameter up to 5 mm) brittle glass borosilicate capsules [9–12]. Pelletier et al. manufactured polyurethane short capsules carrying sodium silicate solution with repair ability [13].

The master concept is graphically illustrated in **Figure 2**. Based on it, numerous studies followed in an attempt to optimize the healing efficiency of concrete systems, in all cases following a series of principal requirements:

- Autonomous healing mechanism should be activated only in the presence of damage. The healing agent should be stored within concrete, protected by a pre-cursor that is strong enough to survive concrete mixing process and in parallel brittle enough to instantly break in tension as crack propagates through it.
- The amount of healing agent should be enough to fill up the cracks formed in concrete under service loads, but should remain limited in volume in order to eliminate agent leaching on exterior surfaces. The agent rheological properties should be adjusted to fulfill this request.

- The healing cycles should be repeatable in order to protect concrete throughout the structure's service life. Concerning the time and spatial spread of damage phenomena on concrete, the self-healing concept should be accompanied by a self-sensing mechanism that detects cracks at any moment in order to control the healing activation procedure.

Section 2 presents the optimized and most promising design strategies: healing agent encapsulation (Section 2.1) and agent delivery vascular networks (Section 2.2). The loading configuration and monitoring techniques used on test campaigns to assess damage and consequent healing are enlisted in Section 3 and a critical review of their effectiveness on damage and healing assessment is given. The combination of the optimal self-healing and self-sensing technologies permits us today to test the feasibility of building constructions with autonomous repair ability. The experimental tests on real-size concrete structures loaded under service loads are reported. This chapter closes with a critical and extensive discussion on healing definition, the transition from laboratory testing to industry and the potential contributions of cutting-edge technologies on future concrete healing concepts.

2. Design strategies

2.1. Encapsulated healing agent

2.1.1. Healing agent

Polymer-based agents similar to the ones traditionally used for external manual injection are used. The preliminary studies discussed the self-healing effectiveness of polymers existed in the market (polyurethane, superglue and epoxy) [10]. It was evident that the polymer chemistry had to be modified in order to optimally perform as encapsulated healing agent:

- The agent should react only with specific catalyst, indicatively air, water or other chemical accelerators.
- The agent should be effectively polymerized enclosed into the aggressive alkaline concrete environment.
- The agent curing should be rapid enough to prevent crack propagation and exposure of reinforcement to ambient environment.
- The agent rheology should permit fast penetration through thin cracks (by means of capillary and gravitational forces), but agent leaching due to low viscosity should be eliminated.
- Optimally, the agent should expand in volume in order to fully seal the crack void.

2.1.2. Agent carrier

Borosilicate glass [12], ceramic [14], cementitious [14], polyurethane [13] and other polymeric [15] capsules were used. Glass effectively breaks in the presence of macro-cracks, however,

cannot survive mixing and long-term exposure to the aggressive alkaline concrete environment. Polymer-based capsules can survive mixing and embedment into concrete, but often do not rupture in the presence of cracks. Finally, ceramic and cementitious capsules appear the most promising. Minnebo and Tsangouri developed the latter capsules by material extrusion [14]. The capsules can survive the concrete mixing and provide the optimal interfacial bonding with the concrete. Enhanced concrete-capsule interfacial bonding ensures instant capsule rupture as cracks form and eliminate debonding effects.

The capsules were initially designed in spherical shape [16], but later the tubular shape [10] was adopted as the optimal shape since it increases the chances of rupture in cracking stage. The size of capsules varies: Yang introduced micro-capsules [16], Van Tittelboom established the short 25–75 mm long tubular capsules and recently longer capsules up to 1 m long were manufactured [14, 17].

2.2. Agent delivery vascular networks

In the recent decade, an alternative effective healing system is under investigation inspired by the blood circulation in vascular networks. Concrete sections are made porous and healing agent circulates through the material providing repeatable repair at different locations [18]. Kuang designed a porous core in concrete and as a proof-of-concept adhesive epoxy was manually injected in the presence of crack [19]. Shape memory alloys and shrinkable polymer tendons were implemented to enhance the effective agent penetration and curing [19, 20]. Upon damage, externally introduced heating activates the tendons which recover and shrink to their original shape, closing in this manner the crack-void and facilitating the bonding at the fractured surfaces. The aforementioned systems provide passive manual healing, but their contribution should not be underestimated since these prototypes introduced the idea of agent delivery vascular networks.

The research team at the Department of Mechanics of Materials and Constructions (MeMC) at Vrije Universiteit Brussel (VUB) worked the past 5 years on the manufacturing of healing agent tubes network that imitates the vascular network flow. A comparative study on optimal tubes composition that considers the material compatibility and brittleness, the bonding conditions at the concrete tubes interface and durability performance reached the conclusion that ceramic and cementitious long (up to 1 m), extruded capsules with inner diameter up to 10 mm optimally perform [14, 17]. The tubes are interconnected by 3D printed connection nodes that often stand below reservoir deposits. The agent is stored in the deposit and is poured into the tubes only in the presence of cracks, procedure manually triggered by an air pressure sensor. Current research investigates how sensor configurations can be implemented into the healing system introducing autonomous sensing of damage and healing process trigger.

In parallel, the on-going research project materials for life focuses on the development of thin tunnels network embedded into concrete carrying organic, but also inorganic healing agent [21]. The tunnels network is built into thin wall elements and its performance under service loads is currently under investigation.

3. Assessment of healing efficiency

The healing efficiency of encapsulation systems is evaluated in the laboratory by performing tests on small and bigger concrete samples that carry the healing system. At first, the intact concrete sample is loaded until a macro-crack is formed. As the crack propagates, it breaks the capsules that pass through, activating eventually the healing process: due to gravitational and capillary forces, the agent is released in the crack void. A loading pause of few hours (often less than 24 hours) follows during which agent polymerization takes place. After the healing pause, loading is applied once again and at this time the response of the healed zone to further cracking is assessed. In the case that numerous capsules are embedded and new cracks form after healing, more than one reloading cycles are applied to detect potential repeatable healing.

In **Figure 3**, different loading configurations and specimen geometries are illustrated giving an overview of the most popular test setups used up to date for healing assessment. Cases 1–3 stand for short concrete beams ($840 \times 100 \times 100$ mm) loaded under 3- and 4-point bending. Only in Case 1, a notch is prepared to guide the crack at the middle section of the beam [22] where limited number of short glass capsules are embedded. Case 2 considers numerous short capsules scattered into concrete and their performance is assessed as multiple cracks form under 4-point bending [23]. Similarly, in Case 3, 4-point bending is applied on a beam that carries a prototype agent delivery tubes network [14]. Cases 4–6 illustrate the tests done on real-size structural elements (beam, slab and wall, respectively). Short glass capsules are embedded along the length of a beam 3 m long and potential multiple cracks healing is assessed under 4-point bending [24, 25]. The prototype tubes vascular network is extended in 2D plane and is embedded into a slab 4 m long and 1 m wide in an attempt to upscale this promising healing system (Case 5, [17]). Finally, the healing mechanisms built in the course of materials for life research project are evaluated by loading and monitoring the cracks formed on 1 m-wide walls as illustrated in Case 6 [21].

3.1. Detection of healing activation

The first step towards autonomous healing is the in time and effective activation of the healing process. The capsules should break in the presence of a macro-crack leading to healing agent

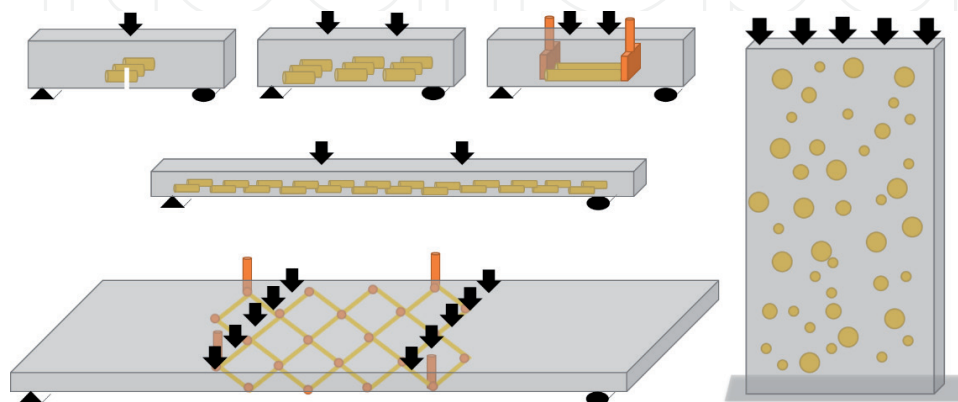


Figure 3. Design and configuration of tests performed to assess the healing efficiency on concrete. Loading is commonly applied in two or more cycles.

release. Continuous monitoring using acoustic emission (AE) is proven to accurately sense the capsules rupture. In **Figure 4a**, the AE hits energy emitted from short concrete beams (design Case 1) is plotted. The beams carry numerous short glass capsules that break as crack forms and propagates. The majority of AE hits are originated from the concrete cracking, however there are series of AE hits instantly emitted carrying energy up to a scale higher than the concrete cracking hits (typically a series consists eight hits captured by the eight sensors attached to the beam). The high energy hits, marked in black in **Figure 4a** are released as the capsules rupture.

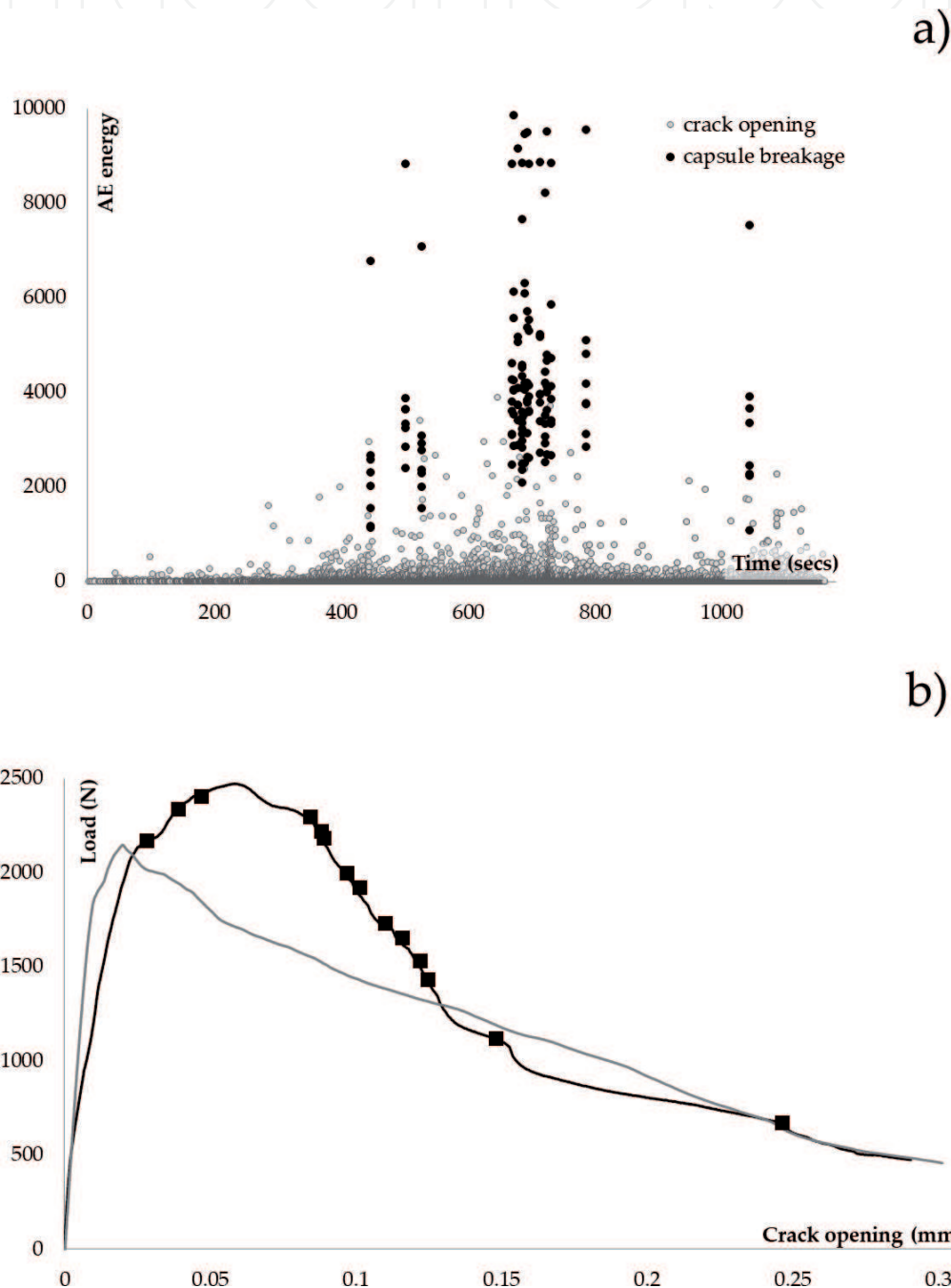


Figure 4. (a) Scatter of AE hits energy where hits emitted due to capsules rupture have values at least a scale greater than the hits originated from concrete cracking; (b) the capsules rupture detected by AE energy analysis are tracked on the load-crack opening curve. The loading curve is compared to reference case at which healing capsules are not used.

AE hits energy-based protocol was used to distinguish the rupture of different types of capsules: glass (**Figure 4a**, [22]), ceramic [14], cementitious [14] and glass with cementitious reinforcement [23]. In all cases, capsule breakages were distinguishable from concrete cracking, however the energy levels drop in the cases that compatible to concrete capsules material is used (i.e. cementitious mortar). In a step further, the capsules rupture can be localized relevant to the fracture stage: in **Figure 4b**, the instants of capsule rupture (detected by AE energy protocol) are plotted along the load-crack opening curve. The AE analysis proves that capsule break in the presence of a macro-crack with size ranging from 40 to 150 μm . Furthermore, the load-crack opening curve of a reference concrete beam (carrying no capsules) is given highlighting the fracture toughness increase in the healing series. Apparently, the capsules enhance the concrete response to fracture and provide an indirect local reinforcement to the structure. The study proves the multi-beneficial contribution of encapsulated agent systems on concrete damage response.

3.2. Sealing efficiency

The healing performance of the proposed technology is variously interpreted. Often in literature, the terms sealing, repair and mechanical restoration are enclosed in the term 'healing'. Effective sealing can be assessed by performing water permeability tests [23], neutron tomography [12] and optical scanning microscopy [26]. The water flow through multiple cracks formed on short and small-scale concrete beams was measured and it was found that only partial sealing can be achieved since the expansive agent only partially covers the crack void [12]. The water permeability configuration loses its accuracy in the presence of several cracks [23]. Scanning microscopy has effectively illustrated the agent release into the crack void. In **Figure 5**, Feiteira et al. visualized the effective sealing of a crack by an elastic polymer agent [26]. In bending after healing, the agent elastically deforms and progressively debonds from the crack faces. The method can provide sealing evidence only if samples of few hundred millimeters are considered, but it loses in accuracy as larger damage zones are inspected [23, 24].

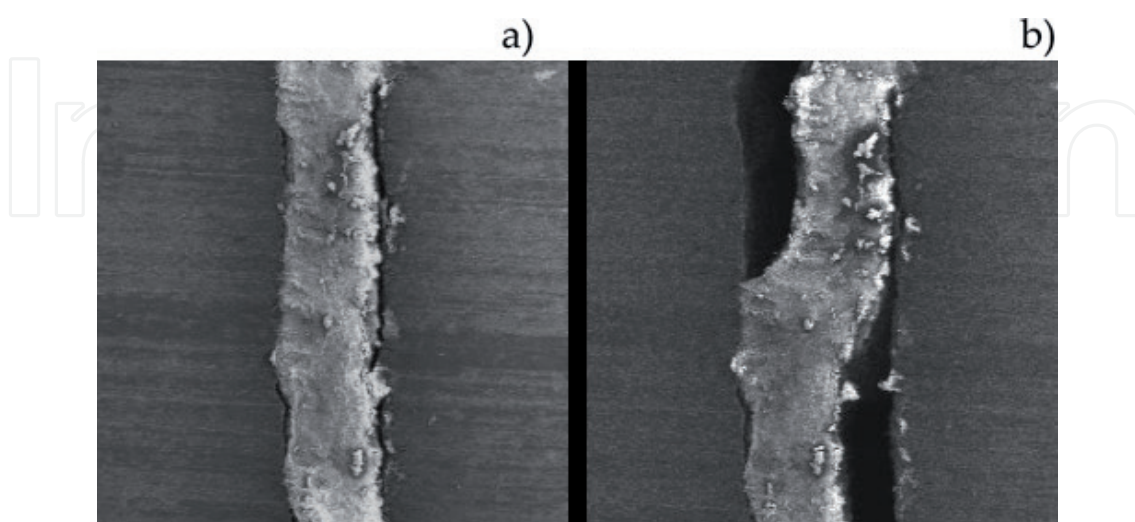


Figure 5. Scanning microscopy: (a) a crack formed on small-scale concrete beam is sealed with an elastic polymer healing agent and (b) at reloading stage, the agent elastically deforms and debonds from the crack faces [26].

3.3. Mechanical reset and repair

Mechanical restoration can be quantified by measuring the stiffness, strength and fracture toughness after healing and compare these values to the ones of intact stage. Extended research was previously performed on short concrete beams that carry a crack control pre-notch and a series of short agent capsules were embedded into them. The analyses have shown that in the ideal case that the unique crack is fully sealed and the agent is well polymerized, stiffness and strength recovery up to 100% can be achieved. In practice and considering an effective healing, typical reset levels are ranging from 40 to 70%.

The mechanical restoration can be associated to ultrasound pulse velocity recovery after healing. In a recent study, a pair of transmitter-receiver sensors is set facing the zone where controlled unique crack will form under load. As shown in **Figure 6a**, the piezoelectric transducers are embedded into concrete beams (design Case 1) during casting, this way eliminating the surface coupling and wave attenuation effects (further setup description can be found in [27]). A damage index that by definition considers both the shift of the arrival time and the amplitude variations is calculated and the outcome is presented in **Figure 6b** for both reference (carrying no healing material) and healing series [27]. It is shown that only in the case of healed samples, the damage index resets at the reloading cycle onset.

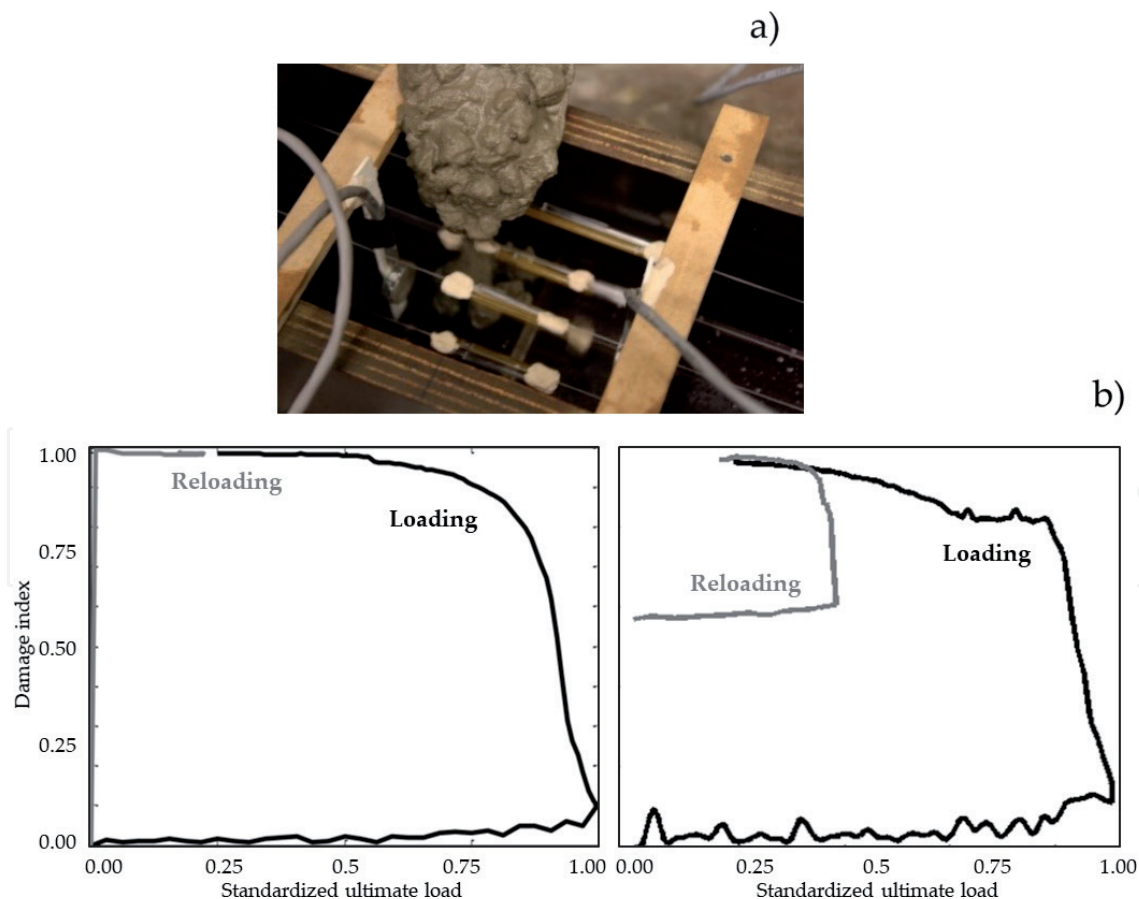


Figure 6. (a) A pair of piezoelectric transducers is detached to the concrete mold. The concrete is cast and the sensors remain embedded into it; (b) damage index evolution at loading/reloading cycles for reference and healing series.

The use of embedded sensors to track damage appears effective monitoring method only when small-scale controlled damage test configurations are considered and significantly loses in accuracy in larger concrete samples (i.e. design Case 4). Monitoring of larger concrete samples requires the development of a well-spread sensors network.

Tracking of repair efficiency appears more challenging as multiple cracks form and interact in concrete (design Case 2). Digital image correlation (DIC) strain concentration mapping can indicate the cracked zones on concrete surface, therefore highlights potential crack closure after healing. In **Figure 7**, a representative cracks pattern is presented. There are five macro-cracks with crack opening up to 300 μm formed on short concrete beams tested under

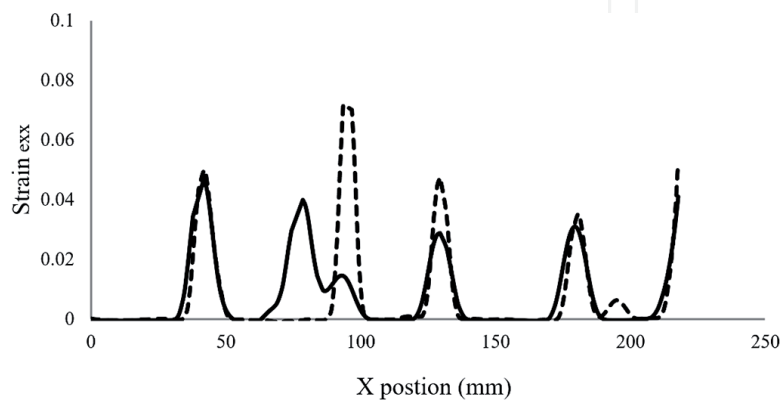


Figure 7. Horizontal strain (parallel to crack opening direction) maps captured by DIC on concrete surface of small-scale concrete beams. One of the cracks formed at the first loading cycle is healed, therefore at reloading stage another crack forms at its vicinity [28].

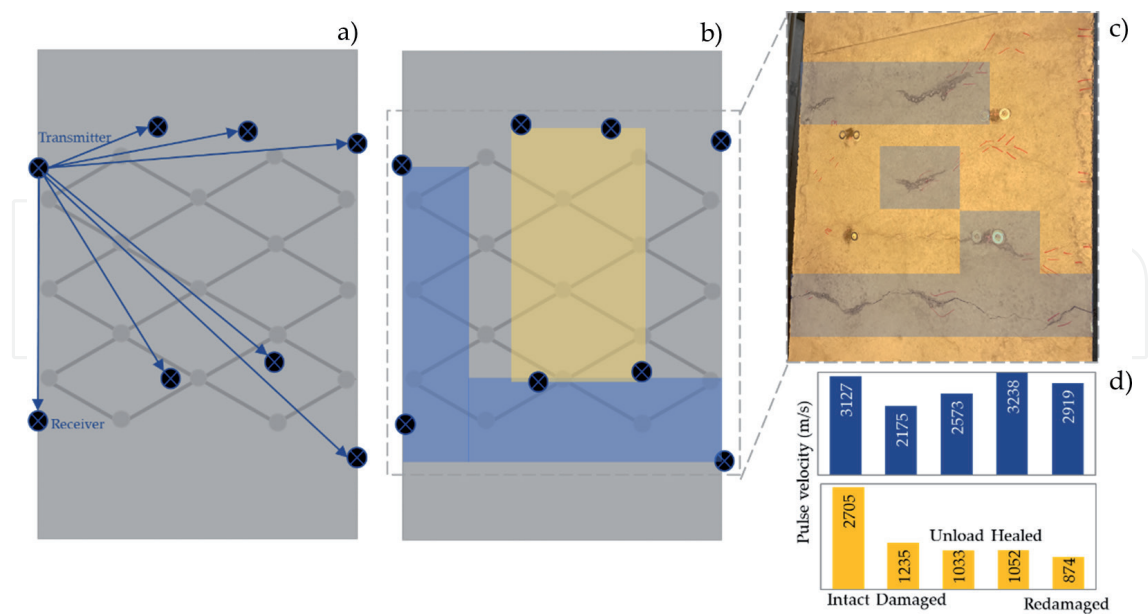


Figure 8. Mapping of ultrasound pulse velocity distribution, indicative of effective healing: (a) transmitter-receivers configuration; (b) maps of healed (blue) and non-healed (yellow) zones; (c) visual inspection of the plate bottom indicates areas where agent is leaching; (d) indicative distribution of velocities for a healed and non-healed zone, respectively.

four-point bending. The majority of cracks reopen at reloading stage after healing, however, one of them is effectively repaired and closed, therefore another crack forms at its surrounding at this second loading stage. The crack closure and opening of new cracks after healing is an evidence of effective material mechanical restoration.

In a recent study, ultrasound pulse velocity is applied to map the structural health condition of a plate (design Case 5) after healing. An array of piezoelectric transducers is attached to concrete surface as illustrated in **Figure 8**. Periodically, each one of the sensors emit a burst pulse (emitter) that travels through the concrete plate and reaches the other seven sensors (receivers). Considering known the spatial distance of the sensors and measuring the pulse arrival time, one can calculate the wave propagation velocity along the transmitter-receiver line. The study has shown that there are zones (marked in blue, **Figure 8b**) where the velocity drops under bending due to cracks formation, but after healing the velocity resets. On the other hand, yellow-marked zones on the plate indicate areas the velocity has not recovered after healing, therefore local healing was not effective. Indicative velocities distribution is illustrated in **Figure 8d**. The healed zones detected by ultrasound pulse velocity measurement are correlated to the zones where agent leaching is detected at the bottom side of the plate (**Figure 8c**), the latter being an indirect evidence of healing activation mechanism.

4. Closing remarks/future perspectives

4.1. Testing protocol establishment

The studies performed to assess the efficiency of newly developed healing technologies are empirically designed. The loading/unloading/reloading testing protocol is well-established since strength, stiffness, toughness or other mechanical feature reset index can be calculated:

$$\text{Repair index (\%)} = \frac{\text{Mechanical feature at intact stage}}{\text{Mechanical feature after damage and healing}} \times 100\% \quad (1)$$

However, there is no standardized test protocol based on which the healing index is obtained. The author proposes the design of concrete elements according to Rilem TC 50-FMC recommendation: small-scale unreinforced concrete beams carrying a pre-notch in the middle section that guides and controls the crack propagation, loaded under three-point bending in a quasi-static mode. Rilem TC 221-SHC should extend its recommendation on self-healing concrete testing protocol.

In this direction, the terms healing, repair, sealing and mechanical feature reset should be studied using different measuring and monitoring methodologies. Indicatively, the water permeability testing quantifies the sealing efficiency, digital image correlation can be used to track cracks repair and load-crack opening curve should be plotted to obtain potential stiffness and strength reset after healing.

4.2. Hyper-sensing in the service of healing

The analytical presentation of the monitoring methods outcome highlights the utmost importance of their application for healing assessment on concrete. An integrated methodology that combines optical, acoustic or other acoustic techniques should be designed providing essential structural health assessment.

4.3. Long-term healing efficiency

The progress on self-healing concrete design is impressive the last decade, still the technology has not reached the market since there is an open discussion on the material sustainability. Up to date, there are no studies done to assess the long-term performance of self-healing systems. It is proven that glass capsules cannot survive the alkaline concrete environment more than few years, therefore their replacement by ceramic or cementitious capsules is advance in this direction. Moreover, there is no evidence of agent degradation as the polymer is stored for several years at the healing reservoir. Life-cycle assessment models should be developed in the near future to predict the potential use of self-healing technology.

4.4. Additive manufacturing of healing systems

The tubes vascular network remained science fiction since the tubes network is characterized by extend geometrical complexity. The recently developed additive manufacturing technology is called to overpass this limitation: 3D printed polyamide plastic agent reservoir and delivery elements were easily and cost-effectively manufactured [14]. A prototype delivery element for a 2D tubes network is presented in **Figure 9a** and **b**. The reservoir is attached to the tubes edges, is externally filled up with agent that is delivered through the tubes only in the presence of cracks. In preliminary studies, the reservoir appears to attract cracks formation due to greater than concrete material stiffness. Ongoing research investigates the potential additive manufacturing of the reservoir by cementitious media.

In plane healing vascular networks as discussed in Section 2.2, the short tubes are interconnected by additive manufactured circular, thin and light-weight PVC nodes (**Figure 9c**) [17]. Holes through which the agent circulates are drilled through the nodes. The tubes are attached to the holes. Additive manufacturing appears an essential tool for the design of real-scale concrete structural elements with healing ability.

4.5. Healing in the service of heritage conservation

Recently the interest on historic buildings conservation has arisen. Following the trend of smart materials of today, the composition of traditional mortars is re-invented in an attempt to optimally repair existed heritage structures. In a recent study, lime mortar casted as historical masonry filler is designed considering commercial crystalline admixtures and tailored encapsulated additives that permit early age autogenous and post-curing autonomous healing [29]. The self-healing lime mortar compatibly fits to the historic mortar and moreover is able to heal micro-cracks and develop superior mechanical capacity. This

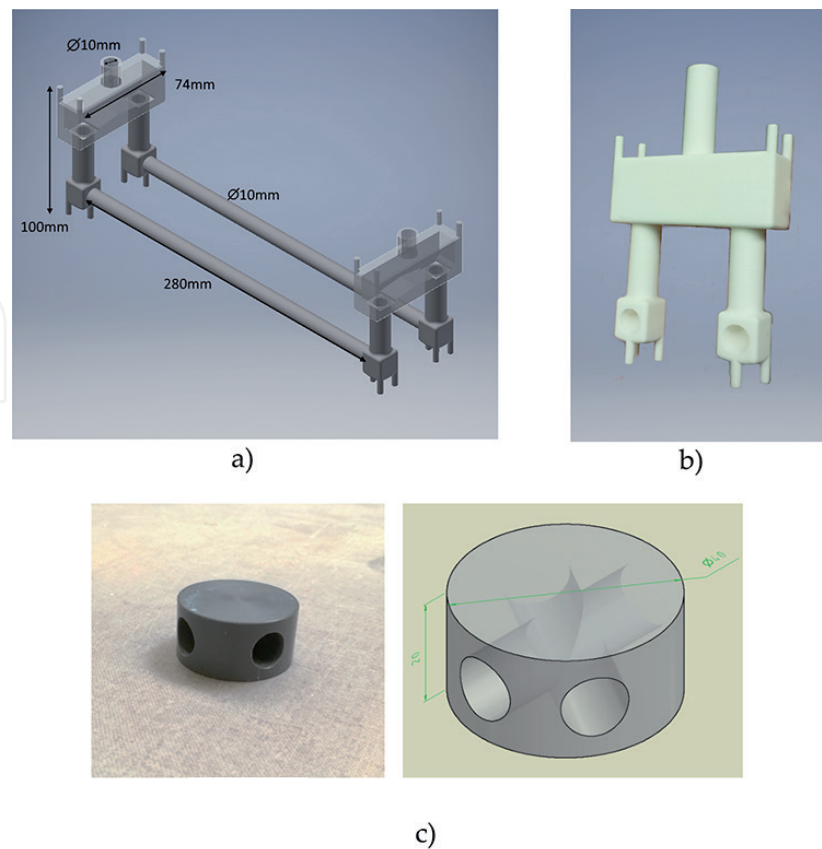


Figure 9. Additive manufactured components of healing agent vascular network: (a) the reservoirs-tubes model (design Case 3 [14]); (b) detail of the reservoir and (c) connection node in slab design case (Case 5 [17]).

pioneering work illustrates the potential application of self-healing technology for the repair and retrofitting of heritage infrastructure, a hot-topic research especially today since the architectural monuments of modernism are tremendously deteriorated and call for effective repair.

5. Conclusions

A retrospective and the current tendencies in self-healing concrete design are presented in this chapter. Self-healing concrete research introduced a new class of intelligent construction materials. The autonomous sensing of fracture and cracks repair feasibility is critically revised leading to the most promising material design: the embedment into concrete of a plane vascular healing agent carrying tubes network. The healing efficiency is assessed using a plethora of inspection experimental methods (AE, DIC, UPV, etc.) and it is concluded that each method contributes to sealing, healing, repair and mechanical restoration characterization. A decade ago, the self-healing concrete idea was abstractly projected in the long future. Recent scientific progress has made revolutionary jumps giving us the confidence that the futuristic self-healing concrete is a product of today.

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Conflict of interest

The author declares that there is no conflict of interest regarding the publication of this chapter.

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