Self-healing concrete—What Is it Good For?

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ABSTRACT: Self-healing of concrete is the process in which the material regenerates itself repairing inner cracks. This process can be produced by autogenous or autonomous healing. Autogenous healing is a natural process, produced by carbonation and/or continuing hydration. Autonomous healing is based on the use of specific agents to produce self-healing, which can be added directly to the concrete matrix, embedded in capsules or introduced through vascular networks. Some examples are superabsorbent polymers, crystalline admixtures, microencapsulated sodium silicate, and bacteria. This review is structured into two parts. The first part is an overview of self-healing concrete that summarises the basic concepts and the main advances produced in the last years. The second part is a critical discussion on the feasibility of self-healing concrete, its possibilities, current weaknesses, and challenges that need to be addressed in the coming years.

KEYWORDS: Concrete; Durability; Microcracking; Transport properties; Mechanical properties.

RESUMEN: Hormigón autosanable - ¿En qué casos es útil? . El autosanado del hormigón es el proceso mediante el cual el material se regenera a sí mismo, reparando fisuras internas. Este proceso se puede producir mediante el sanado autógeno o autónomo. El sanado autógeno es un proceso natural producido por carbonatación y/o por la hidratación continua. El sanado autónomo se basa en el uso de agentes específicos para producir self-healing, que se pueden añadir directamente en la matriz, encapsulados o introducidos mediante redes vasculares. Algunos ejemplos son los polímeros superabsorbentes, aditivos cristalinos, silicato de sodio microencapsulado y bacterias. Esta revisión está estructurada en dos partes. La primera es una visión general sobre el hormigón autosanable que resume los conceptos básicos y los principales avances producidos en los últimos años. La segunda parte es una discusión crítica sobre la viabilidad del hormigón autosanable, sus posibilidades, debilidades actuales y desafíos que deben abordarse en los próximos años.

PALABRAS CLAVE: Hormigón; Durabilidad; Microfisuración; Propiedades de transporte; Propiedades mecánicas.

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1. INTRODUCTION

Reinforced concrete structures are the most used system in buildings and infrastructure constructions. Concrete is an affordable material, easy to produce, which allows variable consistency for application (from dry to self-compacting) and can take different forms and strengths. According to the CEB-FIB Co-de Model 2010, as well as other Codes, the specified design service life of a structure is derived from the requirements given by the stakeholders and the technical implications for structural analysis, maintenance, and quality management. Service life is usually decided depending on the importance of a structure. For common structures, the most usual design service life is 50 years, while in more complex structures design service life is usually increased to more than 100 years. Because of that, materials and construction systems need enough performance to endure these long lifespans, but they also need precise evaluations and knowledge of their long-term mechanical and durability performances, in order to plan realistic maintenance interventions.

In many reinforced concrete structures, cracks are frequent, and they can be acceptable in the structural design as a result of actions considered in the design. In general, concrete elements will suffer combined axial, shear, and bending stresses. These elements will be designed in a way that the compression stresses will be endured by the concrete matrix and the tensile stresses by the reinforcement (Figure 1). The size of acceptable cracks is generally controlled through the material properties, cover, and section design, but mostly through the reinforcement content.

In concrete codes, allowed crack width will depend on the exposure classes, which are chosen depending on the aggressivity of the environment in terms of the risks of corrosion, carbonation, freeze-thaw, erosion, or chemical attacks. In the case of reinforced concrete, values of 0.3 mm of allowed crack width are frequent. In the case of prestressed concrete, cracks of 0.2 mm can be accepted in less aggressive environments. In contrast, no-decompression (and thus, no cracks) is the requirement for the elements under more aggressive conditions. These limits are considered to guaranty that, in the expected service conditions, the structure can maintain its service requirements.

However, it is true that even if the structural conditions are not significantly affected by the crack opening limits allowed for each aggressive class, the durability of concrete and reinforcement can be affected by the mobility of fluids through the open surface cracks. Gaseous materials such as CO₂, water, and acid vapour can be transported even in cracks of up to a few tenths of micrometres (1). Thus, liquids and gasses with aggressive substances can lead to partial deterioration of concrete and corrosion of the reinforcement, affecting the durability and service life of the structure (Figure 1). If cracking overpasses certain limits, either because the design evaluation was wrong or because the expected service conditions were exceeded, deterioration in specific structures could imply high costs for inspection, monitoring, maintenance, and repair.

Self-healing of concrete can be defined as the process in which the material regenerates itself repairing its own cracks, similarly to what happens in some natural materials, such as bones or trees (2, 3). Increasing the self-healing properties of the concrete can lead to mitigate the potential decrease in durability produced by cracking. The immediate objective of self-healing concrete is promoting a partial or total recovery of their physical, mechanical and/or durability properties. Its final objectives are to increase service life or to be able to design more competitive structures. With that purpose, the extent of the properties recovered needs to be quantified accurately as well as the implications in their long-term performance and service life.

Every year, new advances are being researched in order to obtain new ways of producing self-healing in concrete, as well as new methodologies to quantify those improvements produced, in which properties, and under which conditions. Up to now, hundreds of articles have been published related to the self-healing capacity of different types of cement-based materials, including several reviews (4-10). Despite this high number of works published, most of the publications follow a descriptive approach of the results achieved in the different papers, not putting the focus on the questions that still need to be discussed.
This review presents a critical examination of the advances performed in the field of self-healing concrete. The document describes briefly the main processes involved in autogenous and autonomous agents and the methods used for its evaluation to provide a general framework to the reader. Additionally, large-scale tests and the evaluation of long-term improvements have also been reviewed, which are topics of very recent development that are critical to understanding the feasibility of the self-healing systems. Afterwards, the authors critically discuss the levels of efficiency obtained, pointing out some doubts, weak points, and some of the challenges that need to be addressed in the next years.

2. BACKGROUND - BASIC CONCEPTS

Concrete self-healing is the process in which the material regenerates itself repairing inner cracks by an intrinsic (autogenous) or an extrinsic (autonomous) process (7). Thus, self-healing in concrete can be divided into (see also diagram in Figure 2):

- **Autogenous healing**: does not require the use of specific agents added to the matrix and is produced by hydration of unhydrated cementitious material particles or by precipitation of calcium carbonate (CaCO$_3$) (4, 5, 9, 11);
- **Autonomous healing**: produced by reactions of a specific agent intentionally added in the concrete mix design to produce self-healing. This agent can be added directly, embedded in capsules, or introduced through vascular networks. Some examples of the most used systems are superabsorbent polymers (12-15), crystalline admixtures (16-18), microencapsulated sodium silicate (19-21), tubes with adhesives (22-24), and bacteria (10, 25-28).

### 2.1. Autogenous healing

Autogenous healing is a natural process in concrete. The most important mechanisms involved are carbonation of calcium hydroxide (portlandite) and continuing hydration of partially unhydrated cement grains (29). In general, high humidity environments are not able to produce effective autogenous healing, and direct contact with water is required (9, 30-32).

In the case of carbonation, calcium carbonate crystals (CaCO$_3$) precipitate in the crack surfaces due to the chemical reactions between the Ca$^{2+}$ (present in the hydrated concrete matrix) and the CO$_2$ available in the water of the crack. The precipitation of CaCO$_3$ is able to reduce water permeability in cracked concrete elements, with higher healing speed during the first 3-5 days of healing in water immersion (11). However, it has to be noted that some studies (33) reported that the highest concentration of CaCO$_3$ precipitation occurred only at the crack surface.

One factor of significant influence on self-healing is the size of the crack. Smaller cracks have better chances of healing due to the smaller volume that needs to be filled. The maximum crack width reported achieving complete closing through autogenous healing range from up to 0.06 mm (34) or up to 0.20 mm (10, 32, 35) or 0.30 mm after healing during one year (36). Under immersion in CO$_2$-water for 90 days, autogenous healing could heal cracks up to 0.45 mm (37).

Restricting crack width can be used as a method to improve the self-healing of concrete. Two main methods have been analysed in order to improve self-healing: the use of fibres (38) and the application of compression stresses. The type of fibre can also influence the formation of precipitates inside...
the crack (39), and crack control (40). The application of compressive stresses in the element to close the crack and bring the two faces closer also improves self-healing, either if this stress is applied externally (41) or internally through tendons made of shape-memory materials (42).

Continuing hydration is always proposed as an essential mechanism with effects on autogenous healing; however, this process has not received as much attention as carbonation (5). Healing by continuing hydration effect is generally evaluated comparing the response of young matrices with well-hydrated matrices (41, 43, 44), obtaining better healing for younger matrices (41, 43) due to the higher availability of the calcium hydroxide and humidity in the matrix (34). However, the influence of continuing hydration appears to be smaller than that of carbonation. One study isolated continuing hydration from carbonation by working into sealed rooms to avoid the entrance of CO$_2$ in the water (45), and cracks of size up to 15 μm were filled only between 5 and 40%.

One sub-topics that drawn attention is the incorporation of supplementary cementitious materials (SCC) to improve autogenous healing, such as silica fume, fly ash, metakaolin, or blast furnace slag. These materials can improve autogenous healing by taking advantage of their delayed hydration, depending on the remaining reaction capacity of the material at the moment of cracking (43, 46-49). Mixes with higher cement content and/or SCC will have potentially better autogenous healing (18, 35).

All in all, autogenous healing can occur in any type of cement-based material that requires certain conditions to be effective, such as crack widths below 0.15 mm and the presence of water and/or CO$_2$.

2.2. Autonomous healing

Autonomous healing systems use agents engineered to be directly mixed in the cementitious matrix or introduced embedded in an encapsulation system, which protects them until the moment when they are released (50). Their activation can be produced by cracking itself or by the contact with the air or water in the crack, or by other agents introduced into the matrix. If the agent is incorporated in the mix without protection, its efficiency must remain latent until the generation of a crack. If the agent is encapsulated, the capsules must resist the mixing process, and the collision with aggregates or mixer blades (51), and the healing material must have good mobility to ensure an adequate release (9).

This section reviews self-healing agents used to produce autonomous healing in cracks, organised by controlled release of water, inorganic chemical agents, reactive adhesives, and biological agents. Subsequent section 2.3. will shortly describe some encapsulation methods proposed in the literature.

2.2.1. Controlled release of water

The encapsulation of water has been studied to promote autogenous healing in concrete, using absorbent materials that can act as water pockets, such as superabsorbent polymers, vegetal fibres (52, 53), or nanoclays (54). These materials have high water absorption and produce a controlled release afterwards if a crack appears in concrete. The main benefit of these systems is an increase in the self-healing speed compared to autogenous healing. In fact, using cellulose microfibers increased healing rate during the initial days of healing as compared to reference concrete (53), but reaching the same final results. Similarly, vegetal fibres of higher absorption displayed faster healing (52).

Superabsorbent polymers (SAPs) are a particular case that experiences combined effects. SAPs are cross-linked polymers with a high capacity for absorbing fluids (14). When they absorb liquids, they swell to form a soft and insoluble gel (55, 56). Their swelling capacity is highly dependent on their environment (57). SAPs have high absorption on neutral/acid water, but hardly absorb alkaline water in fresh/hardened concrete (56). The estimated absorption capacity of SAPs in relation to its own mass can vary from 500 to 1080 times in distilled water, from 10 to 30 times during the mortar mixing (14, 57, 58). Swelling time also depends on the properties of the SAPs and can vary from seconds to minutes (12, 59).

When SAPs are added into concrete, during the concrete casting and setting, they do not experience a high water absorption nor volume increase because of the concrete high pH. If the matrix cracks and is put in contact with water, two reactions related to self-healing occur:

- Physical blocking produced by the gel: SAPs absorb water and form gels that can fill the cracks. This process can recover water-tightness in the concrete element. This effect is temporary: if the concrete surface is dried, water is released, and SAPs shrink. SAPs are available for reswell if put in contact again with water (55).
- Promoting autogenous healing: The water absorbed in SAPs is gradually released, reacting with unhydrated cement particles and hydration byproducts, promoting autogenous healing. SAPs are added in dry conditions with typical dosages of 0.3-0.6% by the cement weight (13, 14, 58, 59). Dry SAPs can consume part of the water during the mixing process, increasing the plastic viscosity in the fresh state if no additional water is introduced (14). Additions of 1% SAPs by the cement weight can demand an increase in the water/cement ratio from 0.35 to 0.43 (13). The size of SAPs is also an essential factor. Typically, SAPs of size around 500 μm provide good self-healing response, better
than sizes around 200 μm or 80 μm (60), and less reaction time.

One of the drawbacks of SAPs and the additional water to compensate for their absorption is the formation of voids, which reduces compressive strength (58, 59). Another drawback is that SAPs will not work in marine environments, since their swelling capacity is hugely reduced (55, 61).

2.2.2. Inorganic chemical agents

In this group, several types of healing agents have been proposed. Some of these agents can be added without encapsulation, but others need to be encapsulated to be protected until the moment they are required to act. The self-healing agents included in this category are crystalline admixtures, expansive agents and silica-based agents.

Crystalline admixtures (CA) are commercial admixtures that react with the humidity of fresh concrete and with the products of cement hydration, producing non-soluble crystals that promote self-healing of cracks. The term “crystalline admixtures” is a label used in commercial admixtures not necessarily reflecting functionality or molecular structure (9). Some proprietary CA are Penetron Admix and Xypex Admix. CA have been reported to slightly enhance self-healing in terms of crack closing and water tightness (17, 18, 33, 62), and less frequently, also recovery of stiffness and bearing capacity (16). However, some studies did not found recovery of mechanical properties (63, 64). CA are effective only in direct contact with water and for cracks below 0.30 mm (17, 62).

Several expansive additions have been proposed in the literature, such as calcium sulfoaluminate (CSA), (Ca3(AlO2)2(SO4)), magnesium oxide (MgO), calcium oxide (CaO) and calcium sulphate (CaSO4), due to their expansive behaviour. Some authors studied CSA in 10% by the weight of the binder, obtaining better self-healing for cracks up to 0.3 mm (33, 43, 64) in terms of visual closure and water permeability. CSA with a lower amount of CaO and higher Al2O3 and SO3 displayed better results (33, 64). In the case of mechanical properties, only slight improvements were detected.

CA and CSA have been combined in some studies to investigate possible synergies, obtaining improved self-healing (33, 60), with contents of 10% CSA and 1.5% CA by the cement weight, closing cracks up to about 0.4 mm (33). Even though these admixtures are designed to be added directly to the concrete mix, their controlled activation when self-healing is required is a concern. The combined use of SAPs and CA has also been proposed (60, 65), to provide controlled water release for the reactions and showed that CAs produced more self-healing products around SAPs. Similarly, SAPs with CSA agents have also been proposed (60), but further research is still needed.

Magnesium oxide has also been used to promote self-healing due to its expansive properties. Without encapsulation, MgO has been reported to close cracks up to 0.5 mm after 28 days (66), with increasing healing when increasing MgO content. Similarly, bentonite and lime have also been proposed (67, 68), as well as their combinations. MgO encouraged in the formation of brucite and other magnesium hydro-carbonates, while bentonite influenced in the formation of ettringite, and quicklime produced additional portlandite, calcite and calcium-based hydration products (67). Since the addition of MgO can lead to undesired expansions and stresses, its encapsulation has been studied to control its activation (69, 70).

Silica-based agents generally promote the formation of CSH gels, similarly to pozzolanic reactions. The most frequently used agent is sodium silicate (Na2SiO3), but also colloidal silica (mSiO2·nH2O) has been proposed (71). Sodium silicate is one of the most used agents in the self-healing microencapsulated systems. Sodium silicate reacts with hydrated cement pastes in complex interactions with calcium hydroxide (Ca(OH)2), calcium aluminate (CaAl2O4) and non-hydrated C/S/C:S phases to develop hydration products as CSH or CASH (18, 72-74). Most studies that use sodium silicate as a self-healing agent include this product encapsulated, so it reacts after being released out of a microcapsule (20). Microencapsulated sodium silicate has displayed positive impact, healing cracks up to 0.20 mm (75), improving water impermeability through sorptivity (76), and promoting mechanical recovery (21). However, increasing the content of microcapsules can decrease compressive strength (76) because of the increase in porosity. Sodium silicate is also being studied as pore blocking surface treatment (8) to repair structures or to improve water tightness.

2.2.3. Reactive adhesives

Reactive adhesives have been used encapsulated as self-healing systems due to their ability to bond surfaces. Specifically, two types have been used: one-part and multi-component adhesives.

One-part adhesives harden via radiation, heat, or moisture. Because of that, adhesives that harden when exposed to light (77), high temperature (22), or moisture (cyanoacrylates (78, 79) or polyurethanes (80, 81)) have been proposed as healing agents. Once a crack appears in concrete, the interior of the matrix and the adhesive will be exposed to ambient conditions, activating the healing reaction.

Multi-component systems need the addition of two encapsulated elements inside the concrete. Some examples of systems used to produce self-hea-
ling are epoxy resin (82) or one-part adhesives (such as a pre-polymer of polyurethane) combined with an accelerator to improve the reaction (24, 83).

These two systems have the capability of healing large cracks, even up to 0.3-0.5 mm (78, 80), and in several studies, the healed elements even recovered their mechanical properties (80, 83).

2.2.4. Biological agents: bacteria-based self-healing

Microbially-induced calcite precipitation (MICP) has become an area of interest for self-healing of cement-based materials. Bacteria can precipitate CaCO₃ with different metabolic pathways, such as photosynthesis, sulphate reduction, urea hydrolysis or denitrification. For each of these pathways, bacteria also need the appropriate nutrients, which can be, yeast extract, urea, calcium lactate or other compounds. The two processes proposed to introduce self-healing in concrete by MICP are (28): urea hydrolysis by ureolytic bacteria or the respiration process.

Ureolytic bacteria can use carbon sources to produce CO₂ or CO₂⁺, which will react with Ca²⁺ to form bacterial precipitation of CaCO₃. Ureolytic bacteria have the ability to produce a urease enzyme, which hydrolyses urea into ammonia and CO₂, inducing a rapid increase of pH and the precipitation of CaCO₃. The alkali-tolerant ureolytic strains most commonly investigated for their application in cement-based materials are Sporosarcina pasteurii, Sporosarcina ureae, Bacillus sphaericus and Bacillus megaterium (10, 27, 84-86).

In the respiration process, non-ureolytic bacteria act as nucleation sites for the precipitation of CaCO₃ when oxygen is present. The cell wall of bacteria is negatively charged, and, because of that, bacteria can extract cations from the environment, including Ca²⁺ ions that are deposited on the surfaces of the cell wall. Ca²⁺ ions react with CO₂⁺, leading to bacterial precipitation on the cell surface (87). Bacteria are activated and proliferate after the ingress water and oxygen through the cracks. Then, they metabolise organic nutrients (e.g. calcium lactate) instead of urea as the electron donor to produce calcium carbonate (88). The non-ureolytic species most common are Bacillus subtilis, Bacillus cohnii, Bacillus pseudofirmus, Bacillus thuringiensis, Bacillus alkalini-trilicus (85, 86, 88, 89).

Inside concrete, after the formation of cracks, once bacteria are in contact with their nutrients, they are awakened from their hibernation stage (10). Once bacteria are activated, their metabolism starts producing CaCO₃ in the crack. When the crack is completely filled, the bacteria return to hibernation due to the low availability of water or oxygen and the formation of a mineral layer (CaCO₃) covering the bacterial cells. If new cracks form, bacteria will be activated, and the crack will heal again. Therefore, bacteria act like as a catalyst, since they transform a precursor to a suitable filler material (10), but they remain in the matrix.

There are three methods of applying bacteria, each with different efficiency: direct application as spores disperse in the matrix (90, 91), by immobilisation in porous particles like porous aggregates (85, 86, 89), in cellulose microfibers (92) or encapsulation (93). Immobilisation or encapsulation are methods recommended to protect and prolong the life of the bacteria since their survivability is decreased for increasing hydration of concrete (due to the decreasing size of the pores) (10, 94).

Non-ureolytic bacteria are thought to be less efficient than ureolytic bacteria as they do not produce such a rapid increase in pH (95). A non-ureolytic bacteria (86) was reported to heal cracks up to 0.45 mm and to recover 65% of strength. Some ureolytic bacteria also showed good self-healing responses, recovering compressive strength (90, 93) and closing cracks up to 0.85-0.97 mm (93). Bacteria are able to heal larger cracks than autogenous healing (at least two times larger). Bacterial spores for concrete are starting to be commercialised, with few small companies and start-ups providing them, such as Avecom (96, 97) or Basilisk (98), but its commercial availability at competitive prices is still very limited.

2.3. Encapsulation methods

One of the main challenges for achieving self-healing is to protect the healing agents inside the concrete and activate them only at the required moment. With this purpose, several encapsulation techniques are still being developed, either for bacteria or for chemical agents. The encapsulation systems can be comprised by a one-part component embedded (if the healing agent reacts with radiation, heat, water, or air) or multi-component.

The types of encapsulation system can be gathered in two groups: disperse capsules (mostly microcapsules between 20-800 μm (19, 99), but also porous vessels up to 8 mm (69)) and located capsules (mostly glass or ceramic tubes between 10-100 mm length (83, 100, 101)). Both methods need a physical breakage or an increase of the porosity to be activated. The first group is thought to be added to the concrete matrix as an additional compound and is a suitable method for unpredictable or dispersed cracking; while the second needs to be placed in a specific location in a similar way to reinforcement bars, thus being optimal for predictable cracks. The two types of capsules can be filled with different types of healing agents. Figure 3 shows a diagram with a classification of the self-healing agents, introduction methods and their combinations. Previously
published reviews discuss the benefits of different types of microcapsules (4, 9).

An efficient microcapsule should be chosen such that the wall of the microcapsule is strong enough to resist mechanical impacts without breaking during the concrete mixing, with good bond with the matrix and a weak enough shell that it breaks when a crack occurs, and the crack path is not diverged around the capsule (9). Dispersed encapsulation answers to dispersed cracks, so it is more effective for random cracking, but the damage must hit the capsules. Most of the successful studies performed with microcapsules use high amounts (around 4-7% in volume), which can be detrimental to other concrete properties, such as strength.

Located capsules do not need to resist the mixing process since they are tubes or capsules placed in the moulds before pouring the concrete (in a similar way to the reinforcement). However, they still need to endure the impacts produced during pouring. Effective use of located encapsulation to produce self-healing requires crack prediction. This method has high potential, since many authors using adhesive-type products as healing agents encapsulated in tubes achieved excellent recoveries, including mechanical regain (83). However, in some cases, the healing agent released from the tube has been reported as being only a small fraction of the volume embedded (102). The viscosity of the healing agent, the diameter of the tubes, tube slenderness, and the reaction speed will be critical for an adequate release of the agent (83, 102, 103).

Vascular networks are a concept similar to located tubes (104-107), which consists of artificially created channels through which self-healing agents can be pumped inside the concrete element. This system will also answer to predictable cracks but has the advantage of an increase in the amount of material that can be released into the crack. This concept can also be considered a self-healing system even if it is necessary to use a sensor that shows the results to a technician, who would trigger the healing process (108). The difference would lie on the level of intelligence of the system. In contrast, if external material is needed, or the matrix needs to be replaced, the process would not be considered self-healing but repair.

2.4. Evaluation of self-healing

Several methods have been proposed in the last years for the evaluation of self-healing. Besides the specific method used in each research group, some international efforts have been made towards the standardisation, in the context of RILEM committees (TC 221-SHC active from 2005 to 2013), the HealCON project, finished in 2016, and the COST Action SARCOS (started in 2016). However, until date, there are no standards published to test self-healing in concrete.
Different methodologies have been used to evaluate the improvements in properties produced by self-healing. An in-depth review of the methods to evaluate self-healing was published by COST Action Sarcos (7). These evaluation methods can be classified depending on the type of property of interest:

- **Filling of cracks:** Crack filling or closure of a crack is the most straightforward consequence of self-healing, as in the human body is the closure of a wound. The techniques used to evaluate this crack closure include methods to evaluate both, surface cracks, which is the most common method (17, 33, 43, 52) and internal cracks through CT-scans (24, 59).

- **Transport properties:** One of the main objectives of crack healing is to improve the durability of concrete structures. The more important fluids that can enter concrete and that are relevant to durability are water (which may carry aggressive ions) as well as gases, such as carbon dioxide and oxygen. Studies performed in this matter may refer to permeability (flow under a pressure differential) (11, 17, 33, 62, 83, 100), diffusion (flow under a concentration differential) (47, 109), and sorption (flow caused by capillary movement in the pores open to the environment) (12, 76, 110, 111).

- **Mechanical properties:** A complete healing process aims to recover also mechanical properties. Flexural (16, 74, 83) and tensile tests (112) have been proposed with this purpose. Compressive tests have also been used to produce distributed damage (48, 49); however, it is difficult to link directly with a measure of crack healing.

Due to the difficulty of investigating the state of internal cracks, non-destructive tests have also been proposed to evaluate self-healing, since they can give an indirect measure of several properties. Some methods are Resonant Frequency (113-115), Ultrasonic Pulse Velocity (116-118) and Acoustic Emissions (118-120).

The general methodology used to evaluate the effects of self-healing consists of the stages of: creation of controlled damage in the specimens, measurement of properties before healing (e.g. permeability or crack width), healing and evaluation of the properties after healing. In the case of the study of mechanical regain, the damaging stage is also the step of the study of initial properties. Most authors evaluate self-healing by direct comparison of the property of interest before and after healing (23, 62, 64), which can be enough in those cases where healing produces a significant change in the property (such as changing from being permeable to watertight). On the contrary, accompanying reference specimens are recommended when the changes expected in the property of interest are small, in order to distinguish the healing process from hydration or other processes taking place inside the material. Reference specimens can be cracked accompanying specimens stored in an environment where healing is not activated (such as a humidity chamber) or undamaged specimens undergoing the same healing process as the healing specimens (119, 121) (Figure 4).

### 2.5. Moving towards greater challenges

#### 2.5.1. Large scale tests

Laboratory testing is not enough to validate technologies and to introduce them into the construction industry. Most of the tests performed about self-healing concrete in the literature were performed at the laboratory level, and often in pastes or mortars, but not in concrete. This difference can be of particular importance since the percentage in the volume of cementitious materials and self-healing agents are higher in pastes and mortars than in concrete mixes.
However, some research groups worked at the concrete level, and some pilots have been built to evaluate the performance of different self-healing concrete technologies at higher Technology Readiness Levels (TRL).

The first test at a larger scale demonstrating a self-healing technique was performed by the end of the 90s, in real scale models of a bridge deck (22). In that study, four decks were built with different self-healing systems, all based in embedded glass tubes containing adhesive/sealants placed at various fixed locations. In all cases, tubes broke due to the crack formation releasing the self-healing agents. They also detected that after performing additional loadings, an additional release of adhesive occurred and that the adhesives survived for over one year in field conditions.

Self-healing concrete with crystalline admixtures has been used in several projects worldwide in the last decades, such as in Brazil (122): an anti-floating slab (1200 m³ of concrete), a wave-type coverage (320 m³), or one slab in the building basement and the reform of football Stadium complex. In these cases, visual inspections reported no cracks in the hardened concrete. However, no reports about the effectiveness of self-healing were published.

In 2015, a small part of a water channel in Ecuador was the first field self-healing concrete construction using lightweight aggregates containing a bacterial healing agent (123). The channel cross-section had a size of 1×1 m and thickness of 100 mm. Five months after casting, no signs of cracking were detected. In 2016 bacterial self-healing concrete was used in two projects (124): the construction of prefabricated parts of a wastewater purification tank and in two walls of a water reservoir. Monitoring in the following two years suggested that the repairs were effective. According to the authors, in the following time (3 years in the first case and one year in the second) both applications had not yet demonstrated possible benefits of using self-healing concrete for the specific application, but no adverse effects were observed.

Recently, in the context of Materials4Life project, six concrete wall panels of size 1.8 m high and 1 m wide with several self-healing technologies were tested at large-scale laboratory testing and onsite trials (104, 105, 125). The self-healing systems studied were microcapsules with sodium silicate, shape-memory tendons combined with vascular networks, perlite with embedded bacteria combined with vascular networks, and vascular networks. Sodium silicate was the agent chosen to be delivered by the networks. These panels were cracked by applying loads with a hydraulic jack. Crack width and permeability to air were evaluated after cracking and after healing. Their results showed good self-healing performance for the panel with microcapsules with sodium silicate (crack reduction of 60% in 1 month and permeability recovery of 2.5 orders of magnitude in 6 months). The other systems displayed reduced (but potential) healing capability, but the test allowed the researchers to detect those points that will need improvements of the systems in the subsequent studies to optimise their feasibility (125).

In Belgium (126) a slab of an inspection pit was cast with bacterial self-healing concrete. The element has a quadrilateral section in its top view with its sides measuring 0.37 and 0.25 m, and a width of 30 mm. The element also had traditional top and bottom reinforcements with 12 mm rebars. During the inspection, no signs of cracking were detected. Self-healing efficiency was studied in the accompanying specimens for quality control. Over 90% of healing efficiency was reported in terms of crack closing and water permeability. However, no assessment of the self-healing was performed in the full-size structure.

Other pilot actions are currently being built for the H2020 ReSHEALience project. They are structures being of ultra-high durability concrete based on Ultra-High-Performance Fibre-Reinforced Concrete or Textile-Reinforced Concrete to guarantee very tight crack widths, and with crystalline admixtures as self-healing enhancers (127). However, this project aims to monitor durability in cracked conditions, and the differentiation of self-healing expressly may not be guaranteed.

Most constructions that introduced self-healing systems verified self-healing through the lack of cracks, showing the difficulty of evaluating self-healing efficiency in real constructions. This difficulty is mainly produced by the fact that to evaluate self-healing, a crack needs to be developed first. Nevertheless, these pilot activities show promising results to add information in the field behaviour of self-healing concrete.

2.5.2 Service life improvements and life cycle analysis

Self-healing concrete is, in general, more expensive than conventional concrete (some values are discussed in section 3.3). Therefore, it should present benefits that justify its use in structures, such as improved performance, reducing maintenance costs and/or increasing the service life of the structure. Several factors have to be considered to estimate the long-term behaviour and benefits of self-healing concrete in comparison to conventional concrete. Some of these factors are the time needed for the self-healing reaction, time that the self-healing agent will remain active, and its repeatability in the long term, the response in large-scale elements, etcetera.

The improvements in long-term behaviour (mechanical and durability properties) have to be investigated to verify if the extension of service life produced by self-healing is worth the inversion. That
quantification is usually based on models on chemical and fluid transport processes, but also on specific aspects that are of interest when modelling self-healing, such as breakage and release of the healing agents in the different systems (128).

One recent publication investigated the service life improvement produced by the reduction of chloride diffusion generated by self-healing (129), using as a basis a modified version of Fick’s second law of diffusion. This study reported that the application of self-healing concrete with polyurethane encapsulated in embedded glass capsules could reduce the chloride concentration in a cracked zone by 75%. As a consequence of this, the service life of steel-reinforced concrete slabs in marine environments could amount to 60-94 years, as opposed to 7 years for ordinary cracked concrete. Another work with crystalline admixture (2% of cement weight) in concrete obtained a reduction up to 30% in chloride ion penetration, which could increase up to 34% the structure service life (130).

Another step to analyse the benefits of using self-healing concrete would be to perform life cycle assessments (LCA). In (131) a self-healing slab made of engineered cementitious composites (ECC) with SAPs was compared to a traditional solution of steel-reinforced concrete slab suffering 300 μm wide and 25 mm deep cracks, and its associated repair actions, located in exposure class XS2. Their cradle-to-gate life cycle assessment showed that the self-healing slab reported lower impacts compared to those of a traditional concrete slab considering the required cover replacements. Similarly, LCA calculations in (129) indicated 56%-75% of environmental benefits when using encapsulated polyurethane as compared to ordinary cracked concrete. The reduction of repair actions principally produced these benefits.

Despite the promising results of these studies, the publications that cover this topic are only a few, and all of them are very recent. The authors believe that more research and discussion is needed to have a factual basis for the quantification of the potential long-term improvements that can be obtained through self-healing concrete.

3. CRITICAL DISCUSSION ABOUT THE CURRENT SITUATION

Self-healing concrete is a topic in continuous improvement. Many investigations have sought to understand and enhance the autogenous capacity of concrete and to design new techniques to achieve this property through autonomous healing agents.

Ideally, a complete self-healing material would be able to heal itself infinite times and to recover its initial properties perfectly (132). Van der Zwaag also indicates that current materials are closer to “minimal” self-healing materials than to “complete” or ideal self-healing materials. An “ideal” self-healing material not only should heal completely damage of any size, every time it is needed, it should also have similar or superior properties to current materials.

In the case of concrete, most self-healing designs can be considered to be at the halfway point between ideal and minimal self-healing materials (probably closer to minimal). Thus, it would be necessary to evaluate the convenience of the additional initial cost or to accept future repairs.

In this section, the authors expose a critical discussion, regarding the maturity level of the self-healing technologies for concrete as well as discussing, which elements and situations have interesting potential.

3.1. Maturity level of the technology

Technology readiness level (TRL) is an indicator for estimating the maturity of technologies and follows a numerical scale from 1 (lowest score) to 9 (higher score).

In the case of self-healing concrete, most of the developed techniques reported in the literature were proven by experimental tests under ideal laboratory conditions (TRL 4) and some of them under relevant environments (TRL 5). Only a few technologies have been tested in upscaled tests, that is, pilots or demonstrators, with TRL levels between 6-7 depending on if the environment is relevant (TRL 6) or operating (TRL 7). Higher TRLs correspond to the commercial levels; these levels are not reached in the field of self-healing concrete. Significant research has also been performed in TRL levels 1-3, to test and prove concepts, and studies at this basic level should still be promoted to verify novel ideas or technologies that produce a more efficient self-healing concrete. The authors are considering as a requirement of a system to reach TRL 8-9 to have used its self-healing properties in an operational structure successfully. In the case of methods to evaluate self-healing, high TRL would mean that there are international standards that can be implemented in some labs and that construction sites could hire laboratories for a report using standards.

Figure 5 displays the TRL ranges of different technologies related to self-healing concrete, as interpreted by the authors, which has been performed for scientific discussion purposes.

Regarding the self-healing agents:

- Cement and Supplementary Cementitious Materials have demonstrated in operational environments that autogenous healing exists and, under certain conditions, seal cracks (9, 44). However, they have not been used on purpose to produce controlled self-healing.
- CA are commercially available and have shown their potential as waterproofing admixture (de-
pending on the mix) (133), with some criticisms due to the extent of the improvements (134), and has been used in several constructions. Improving self-healing is still in a lower TRL level, currently validated in relevant environments (16-18).

- Bacteria concept has been validated in laboratory and field pilots (123, 124), but the products are not generally produced at large scales (96, 97). Currently, bacteria production is limited, which complicates its application in concrete structures, but its production may improve in the next years.

- Encapsulated silicates (19-21) and adhesives (22-24) have shown excellent results in onsite trials (104, 105, 125). However, these agents are usually prepared in the laboratory (81), and in the case of being produced industrially, they are still at developing stages. Therefore, no commercial information is readily available, and their feasibility in the construction field is very limited.

- SAPs have been validated in laboratory conditions (12-15); however, no results are available in large scale elements yet.

Regarding the introduction methods, agents of direct incorporation (CA in (122)), in porous aggregates (123), and, very recently inside glass tubes (126), have been used in operational environments. Therefore, the efficiency of self-healing in this point has not been enough evaluated. The rest of the systems reach the level of system demonstration in relevant environments since they have been tested in pilots (104, 105, 125, 127).

Regarding the evaluation properties, there are no standards, and in full-scale constructions, self-healing has validated only visually (122, 123). At laboratory and pilot conditions (104, 105, 125, 127), some transport properties and mechanical tests have been validated. Six interlaboratory testing programs are being developed in the framework of the COST Action SARCOS. One program has already been finished (135), and the six laboratories involved obtained comparable sealing efficiencies, highlighting the potential of the methods used for further standardisation. However, further research is needed to obtain standard methods as well as accurate theoretical models, and, in a later stage, to evaluate the improved expected life span.

### 3.2. What is self-healing concrete good for?

In this section, the authors discuss several aspects of interest to discuss the potential of self-healing concrete, highlighting aspects that are still missing in current developments.

a) In what type of concrete element?

In the opinion of the authors, the use of self-healing concrete conceptually makes much sense for reinforced or prestressed concrete, since reinforcement is needed where the concrete matrix may work in cracked conditions. Self-healing of these cracks may protect from the entrance of aggressive agents, such as water or chlorides, towards the reinforcement or delaying carbonation (and thus, de-passivation). Structures with water-tightness requirements are also a potential niche for self-healing concrete since it could ensure the functional requirements of the structure (4, 136).
Due to the importance of durability in reinforced concrete structures, some structures in aggressive exposure conditions are dimensioned following the condition of crack control for durability purposes instead of fulfilling the mechanical requirements solely. This condition represents an over-cost in the structure and could even represent an additional constructive difficulty. Therefore, there would be a potential benefit if self-healing concrete guarantees an improvement in durability.

b) For cracks at what construction stage?

Cracks can be produced in concrete during execution, during service conditions or after suffering accidental actions.

At the execution stage, adverse exposure conditions of the element and inadequate curing may produce cracks, such as those produced by shrinkage. A market study (137) indicated the presence of cracks was the main problem reported in constructions, with a total of 90% of the cases. Additionally, it reports that they considered poor execution to have led to cracking. This study also reports that in 73% of the cases, the problems were caused by water ingress. Unexpected construction cracks should be avoided entirely. The authors consider that investing in a better quality of the element and the construction process will be more cost-effective than investing in self-healing concrete as a system to avoid construction cracks.

In the case of concrete structures working under service conditions, codes on the design of reinforced concrete structures stipulate a maximum allowable width for surface cracks. These allowed cracks can have a range between 0.1 and 0.3 mm depending on the environment and the structure. The range of cracks is where most self-healing systems can produce efficient healing (17, 33, 43, 62, 64, 75, 78, 80) and then, where most of the potential impact can be produced.

In some applications, cracks are predictable in location and difficult or expensive to eliminate. This situation happens in half-joints in precast concrete structures, or in certain elements sensitive to shrinkage. In this case, the authors believe that the use of located macro capsules can be a good alternative to increasing reinforcement content for some cases. This case was explored in the bridge deck prototype cracked by shrinkage, which had embedded tubes and reported the sealing of the crack (22). When diffuse cracking is expected, and their location is not predictable, technologies based on distributed products (such as microcapsules or CA) would be more efficient.

Self-healing systems designed to heal damage produced by accidental or not frequent actions are difficult to justify economically and conceptually. In structures that experienced extreme actions that can be under extremely damaged conditions, repairing the element usually is not a priority, because the focus is usually given to maintain the stability of the structure during enough time to ensure people's safety. Afterwards, these structures are repaired or retrofitted, but sometimes, the solution chosen is demolition and reconstruction of the damaged element.

c) For which environment?

Some self-healing agents do not work effectively under certain environments, such as SAPs in marine environments (55, 61) or CA in environments without direct contact with water (17). Systems based on reactive adhesives can activate with different systems, moisture (78, 79), light (77), high temperature (22), or with their activator component (24, 83). For those self-healing systems that are activated with water, the presence of water can become at the same time a mechanism that transports aggressive agents that start some degradation phenomena as well as the activator for self-healing reactions, in a kind of love-hate relationship (138). Each self-healing system has different optimal conditions for their reactions, and the system should be chosen depending on the specific situation.

d) What type of recovery?

Crack closing and the recovery of water tightness has been widely reported, such as in (11, 17, 18, 33, 110). However, consistent recovery of mechanical properties has only been reported when using embedded tubes with adhesives as self-healing agents, such as in (81, 83). It should be mentioned that visual closing does not imply necessarily improvements in other properties like durability or mechanical recovery (33).

Reinforced concrete structures are designed considering cracked conditions in the service state; in fact, deflections are evaluated considering the cracked concrete stiffness. The contribution of concrete tensile strength is usually neglected and is only considered through its influence on tension stiffening. Given this premise, self-healing concrete thinking on the mechanical recovery of the cracked zones does not seem a promising concept. The authors consider that the highest potential for self-healing technologies is to recover durability-related properties. However, mechanical properties could be of high interest with the purpose of controlling crack opening rather than the purpose of mechanical regain of the structure itself.

Water tightness recovery may produce a reduction in the rate of deterioration of concrete and the reinforcement, and to produce an increase in the service life of the structure. Only a few studies are quantifying the improvements in durability and service life, such as (129, 130). However, in the opinion of the authors, this is a key point, and improvements in terms of long-term behaviour need to be investigated to verify if the extension of service life produced by self-healing is worth the money inversion.
e) In-depth formation of the healing by-product and bond with the matrix

Some points related with the formation of the self-healing by-product are not enough developed in the opinion of the authors: such achieving self-healing in-depth, characterisation of the properties of the healing by-product (compacity, density, brittleness) and the bond between the healing by-product and the concrete matrix.

Several studies suggest that autogenous healing and some healing promoters are producing only surface blocking (33, 139). It should be mentioned here that most studies reporting crack closing were performed analysing surface cracks. If complete self-healing is achieved in all the crack depth, it will be more likely that blocking of transport agents through the crack will be more effective. Similarly, healing by-products of higher density will provide better protection from the entrance of aggressive agents. Some options that could happen in the self-healing process of a crack are displayed in Figure 6.

A strong bond between both crack faces and the filling materials will allow the distribution of stresses in a larger contact area. Thus, assessing and improving the bond between the old matrix and the filling material would be a potential improvement step to obtain consistent durability improvements as well as a preliminary step before being able to recover effectively mechanical properties either completely or partially.

Another goal for self-healing concrete is to keep the self-healing products attached to the crack wall to maintain the crack closed. If the filling material can endure cracks’ movements (due to repeated loads or new actions), self-healing will be more efficient. To obtain this, healing products that allow plastic deformation may produce a benefit over rigid products.

f) Can current limits of crack opening be changed?

Crack limits stipulated in reinforced concrete structure codes depend on the environmental condition of exposure and the cover of the primary reinforcement. The size of allowed crack widths can reach up to 0.10 mm for highly aggressive environments, and up to 0.30 mm for non-aggressive environments. This limit is more restrictive for prestressed concrete, not allowing cracks nor decompression of the element.

In an element loaded with combined flexural and compression stresses, the crack will be developed in the tensioned layer and will be V-shaped, with the maximum opening in the most tensioned surface (1). When the codes limit values of allowable crack width, the value is established assuming that at the reinforcement level, crack width will be even more reduced. For example, a depth of penetration of about 1 mm can be assumed for a crack with a surface opening of 10 μm (1). There are still some unclear points regarding crack propagation and its internal geometry as well as regarding the durability degradation produced in concrete with small micro-cracks. Length, depth of penetration, density of microcracks and interconnectivity are other crack parameters considered of relevance that have not been widely studied in the literature (1).

What is clear is that with the presence cracks, it is a matter of time that concrete suffers the degradation produced by the aggressive agents that access the matrix. If these cracks reach the reinforcement level, corrosion of the reinforcement will be accelerated. Therefore, it is essential to control the formation and propagation of cracks and, if possible, heal them as quickly as possible. In this way, the velocity of the healing reaction becomes of high importance.

A typical value for obtaining efficient self-healing effectiveness is one month if produced by mineral additions or crystalline admixtures (17, 33). During that time, no significant amount of aggressive agents should have entered the cracked matrix (neither to have reached the reinforcement). The velocity of the reaction is one of the critical advantages of using some resins as a self-healing agent, which in a matter of hours can seal the cracks efficiently (80, 82), or SAPs (12, 59), which expand in few minutes.

Successful self-healing concrete designs open a discussion on the current limits of crack opening. Since self-healing could contribute to the closure of these cracks the codes could be more tolerant in situations of high environmental aggressiveness;
or stipulate a longer minimum service life for the structures, if the current limits of crack opening are maintained. Nevertheless, these changes can only be considered if self-healing systems demonstrate their capabilities to improve the durability of cracked elements in the conditions of interest.

3.3. Commercial situation and potential applications

Currently, the price of self-healing agents can vary a lot depending on several factors, mostly due to the novelty of the topic. The price of bacteria to produce self-healing concrete can vary between 714-5760 € per m³ of concrete (97) depending on the production process. The price of crystalline admixtures in the recommended dosages varies between 50-100 € per m³ of concrete since no additional workforce is needed. These values may change substantially in the next years due to the expected new developments and improvements in production techniques.

The cost of self-healing concrete compared to the ordinary concrete is still high given their effectivity, a fact that has limited its application in civil constructions. Using self-healing would reduce the frequency and cost of maintenance during its life cycle, the need for monitoring, inspection, and repair of structures. That would promote greater sustainability, because of the fewer interventions, material resources and energy used, and lower emission of pollutants (140).

One weakness of several methods that produce self-healing is that they also produce a decrease of compressive strength due to the introduction of voids, such as using SAPs (58, 59) or microcapsules (141-143). This point should be considered since different contents of these self-healing materials will produce a different extent of self-healing, but also different decreases of compressive strength, and thus, a compromise needs to be reached fitted for each case.

Figure 7 shows a decision-making diagram to evaluate if the use of self-healing systems is of interest for solving a concrete construction problem considering the aforementioned aspects. Some of the questions that the constructor would need to consider are, “What is more cost-effective to use self-healing concrete or…”

- to improve the quality of a concrete system (such as better-quality concrete or appropriate joints in pavements)?
- to increase the amount of reinforcement (to better control cracks)?
- to repair the system once a certain damage threshold has been reached?”

A proper study of these alternatives needs a complete analysis, such as a cradle-to-grave analysis, to compare the cost of each alternative.

Current development and costs suggest that the application of self-healing concrete would be justified now only in high demanding applications (144), like tunnels and marine structures, where safety is a major problem, or in structures where the accessibility for repair and maintenance is limited. Other structures with a strict level of safety that may experience some benefits are some bridges, hydroelectric dams, buried reservoirs, retaining walls, raft foundations in contact with water containing chlorides or sulphates. Additionally, in prefabricated elements self-healing can also be beneficial to heal cracks produced due to accelerated production or to heal expected cracks in located points.

Due to the current high cost or low-moderate performance, self-healing concrete seems to be not justified in other structures such as small prefabricated elements or structures in low humidity environments. However, most of the current systems and materials for self-healing concrete are still young technologies, and a significant advance is expected to be achieved in the next years. Not only more efficient self-healing technologies are needed, but also standardisation in methodologies to evaluate self-healing efficiency will be useful to ensure a fair comparison of self-healing agents. All the advances produced in these points, and others mentioned in this review, will allow self-healing technologies to be progressively introduced in the construction field.

4. CONCLUSIONS

Autogenous healing of concrete is a natural phenomenon that heals very small cracks (under 0.15 mm), and that is produced by continuing hydration and carbonation. Autogenous healing can be enhanced with specific concrete compositions and by the introduction of supplementary cementitious materials, such as pozzolanas. Autonomous healing of concrete is based on the introduction of specific agents to produce self-healing. The most studied autonomous agents are SAPs, CA, encapsulated sodium silicate or adhesives, and bacteria. Each of these agents has a different functioning basis and different effectiveness under certain environmental conditions.

Dispersed agents have been reported to heal cracks with a small opening (especially at early ages), generally up to 0.30 mm. Cracks larger than 0.3 mm can only be healed efficiently by embedded tubes with adhesives in located positions and bacteria. These two latter systems are also those that showed higher efficiencies, being able to recover some transport and mechanical properties in several studies.
There are no standardised methods to assess the effects of healing on concrete. The methods used in the literature to assess the crack closing are widespread. However, they do not always show the level of internal healing nor the efficiency of the filling products against durability. Techniques for assessing permeability, diffusion, absorption in cracked conditions, as well as the recovery of mechanical properties, are still being discussed. In this sense, the standardisation of the methods to evaluate the self-healing effects is a great task that must be overcome in the near future.

There are still some challenges to overcome, such as producing some healing agents at an industrial scale or the evaluation of self-healing in a structure in operating conditions. Self-healing of concrete is still practically in laboratory scale, except for some remarkable reduced-scale pilots. These pilots demonstrated the potential of self-healing concrete, especially in the case of microencapsulated sodium silicate. Some self-healing agents have been used in high-volume constructions, especially CA, and bacteria. However, their self-healing effectiveness was not verified, and only the absence of cracks was verified. The authors believe that more experimental tests in larger-scale elements are necessary to evaluate the self-healing capabilities better, mainly thinking in durability properties, and covering the most diverse environmental conditions.

In the authors’ opinion, the use of self-healing concrete has potential for reinforced structural concrete elements, where the concrete matrix works under cracked conditions. Self-healing of these cracks can produce recovery of water tightness and protect against the entrance of aggressive agents such as chlorides, sulphates, or CO₂. This increased protection can improve the service life of the concrete structure, and thus, self-healing has a high potential in terms of durability recovery. Once enough efficiency is demonstrated, they could even modify allowable crack widths from the concrete structural codes.

Nowadays, self-healing concrete has a higher cost than conventional concrete, and its application is justified only in cases with high safety requirements, such as tunnels or marine structures, or in structures where accessibility for repair and maintenance is limited. However, their potential applications are likely to be widened in the next years, as long as self-healing agents are improved and developed at larger industrial scales. If the weak points that have been discussed throughout this review, were successfully developed in the upcoming years, self-healing systems could be a pillar for obtaining more durable reinforced concrete structures.

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