# Encapsulation Technology and Techniques in Self-Healing Concrete

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**Abstract:** Throughout concrete structures' service life span, deterioration inevitably occurs. A typical phenomenon of deterioration in concrete structures is cracking, which affects durability and integrity of these structures. Repair and maintenance of concrete structures are labor and capital intensive; it can also be difficult to access the degree of damage after the construction is completed. Self-healing is a possible solution. An encapsulation strategy is widely considered as a versatile and effective strategy for self-healing. In this review, attention is focused one valuation of different healing agents and encapsulation techniques. Eight key factors that affect the effectiveness of self-healing by encapsulation are discussed; these are (1) robustness during mixing, (2) probability of cracks encountering the capsules, (3) curing time and condition, (4) effect of empty capsules on concrete strength, (5) controllability of release of healing agent, (6) stability of healing agent, (7) sealing ability and recovery of durability and strength of concrete matrix (as a result of self-healing), and (8) repeatability of self-healing action. Finally, gaps in current research and important areas for future research are identified. **DOI: 10.1061/(ASCE)MT.1943-5533.0001687.** *© 2016 American Society of Civil Engineers.*

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## Introduction

It is well known that one of the weaknesses of concrete is its vulnerability to cracking. Cracks may occur when concrete is in a plastic state or after it has completely hardened. Concrete may crack due to plastic shrinkage, thermal stresses, settlement, drying shrinkage, weathering, corrosion of reinforcement, or due to applied loading (ACI 2007; Fernandez 2012). Cracking may also be due to coupled action of two or more of these factors. For example, shallow cracks caused by plastic shrinkage or drying shrinkage may propagate under externally applied loading at a much lower stress level than the concrete is designed for. Concrete has low tensile strength and therefore concrete constructions are often combined with various types of reinforcement to resist tensile stresses. Although this is a measure to control concrete cracking, complete crack prevention is almost impossible. For example, reinforcement act as local restraint against free shrinkage when concrete is in a plastic state, causing settlement cracks to form (Dakhil et al. 1975). Initially, microcracks may be formed, which propagate under external loading or other factors causing loss of structural integrity. Microcracks in concrete also affect durability by allowing ingress of corrosive substances into the concrete matrix, which lead to corrosion of steel and loss in tensile strength. Such occurrence may lead to more adverse problems such as spalling and even premature structural failure. Therefore, maintenance and repair are necessary to seal cracks to reduce permeability and restore durability of the structure. However, in some cases, manual repair works are difficult because cracks are invisible or their locations cannot be easily accessed.

In Europe, half of annual construction budgets are allocated to repair works (Cailleux and Pollet 2009). Van Breugel (2007) opined that "enhancing the longevity of our built infrastructure will undoubtedly reduce the impact of mankind's activities on the stability of the biosphere." This implies that extending the service life of existing infrastructure can reduce the demand for new infrastructure, cost, and pollution.

Damage management is an alternative concept to damage prevention, and is based on the principle that damage in structures is tolerable as long as it is healed or can be rectified in time (van der Zwaag 2007). One such damage-management concept is self-healing concrete. This concept is largely inspired from the wound-healing mechanism of human body, which can heal itself up to a certain level of damage by releasing biological agents to the wounded spots. In general, there are three main approaches of self-healing. These are elaborated in the next section.

## Three Approaches to Self-Healing

The three broad approaches to self-healing in concrete, as mentioned by Van Tittelboom and De Belie (2013), are autogenous, vascular, and capsule-based self-healing.

### *Autogenous Self-Healing*

Autogenous self-healing relies on the composition of concrete and is accomplished by hydration reaction of cementitous products within the matrix, or by reaction of polymeric substances in the matrix. Autogenous self-healing has been widely studied (e.g., Neville 2002; Ramm and Biscoping 1998; Yang et al. 2009; Jacobsen et al. 1996; Granger et al. 2007; Li and Li 2011).One of the weaknesses of this approach is the limitation posed by crack width. Autogenous healing is primarily effective for very narrow cracks; this observation was supported by TerHeide and Schlangen (2007), TerHeide et al. (2005), and TerHeide (2005). Specifically,

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different researchers studied the effectiveness of this technique in sealing cracks of different widths, of 5–10 $\mu$m (Jacobsen and Sellevold 1996; Sahmaran et al. 2008), 200 $\mu$m (Edvardsen 1999), and 300 $\mu$m (Clear 1985). It has been acknowledged that wider cracks that are detrimental to durability of concrete structures cannot be effectively healed by autogenous healing. This limitation was addressed by Li and Li (2011), who proposed using engineered cementitous composites (ECC) containing synthetic fibers, such as polypropylene (PP) and polyvinyl alcohol (PVA), to restrain crack width.

Furthermore, a constant supply of water must be present to support the hydration process so that cracks can be completely sealed. Therefore, the phenomenon of autogenous healing may be more prominent in fresh or young concrete (Van Tittelboom and De Belie 2013), whereas carbonate precipitation may be the pronounced mechanism at later stage (Neville 2002). In order to accommodate more water for further hydration, attempts were made to include superabsorbent polymers (SAP) in the concrete mixture (Lee et al. 2010; Kim and Schlangen 2011; Snoecket al. 2013), which can store and supply moisture over longer period of time. However, when water is released from SAP, pores or voids are formed in concrete that become weak links in the matrix. Cracks may even propagate through them during the service life of the structure (Lee et al. 2010; Snoeck et al. 2013).

Several researchers (Termkhajornkit et al. 2009; Sahmaran et al. 2008; Na et al. 2012) attempted at using supplementary cementitous materials, such as fly ash and blast furnace slag, to stimulate autogenous healing. Materials like fly ash and slag hydrate at slower rate than cement, and therefore, unhydrated particles of such minerals promote autogenous healing at later stage of concrete. However, the disadvantage of this approach is that the healing agent is consumed in the process and may not be available for further hydration at later stage. Addition of crystalline additives and expansive agents, including calcium sulphoaluminate, may heal cracks by expansive reaction (Sisomphon et al. 2011b). However, some cracks may form in the matrix due to expansion during healing.

Jonkers (2007) proposed the concept of using bacteria spores to mediate the healing process by precipitation of calcium carbonate. However, in their preliminary research (Jonkers 2007; Jonkers and Schlangen 2009; Jonkers et al. 2010), although the bacteria initiated the precipitation and deposition of calcium carbonate at the crack faces when added to the fresh concrete mix, they did not survive for a long period due to two reasons: the strongly alkaline environment of the concrete mixture and the shrinkage of pores due to cement hydration.

In summary, autogenous healing mechanism has several inherent weaknesses, including (1) dependency on age of concrete; (2) need for a long-lasting internal source of water; (3) survival of bacteria for carbonate precipitation; and (4) need for limitation on the width of the crack that can be healed.

### Vascular Self-Healing

The vascular approach of self-healing closely mimics the vascular network system in the human body. A network of tubes can be installed in concrete to deliver a healing agent to the cracked/damaged sites. In this approach, healing agents are confined in hollow tubes or network of tubes and supplied by an external source. There are two means of achieving self-healing by vascular approach: single-channel and multiple-channel systems. When only a single-component healing agent is used, the single-channel vascular approach is used; when it involves healing by the reaction of two healing agents, multiple channels are used.

Although external supply of healing agent is effective, technically it is not self-healing per se, since it requires external intervention. Moreover, although feasible at laboratory scale, it is difficult to cast concrete with a network of pipes for vascular self-healing on actual construction sites.

In many ways, the problems encountered by these two aforementioned methods can be addressed by capsule-based self-healing.

### Capsule-Based Self-Healing

In capsule-based self-healing, the capsules provide mechanical protection to the healing agents and only release them after being triggered by cracks (by capsule rupture or diffusion), moisture, air, or a change in pH of the pore solution in the matrix. In cases where cracking is the trigger mechanism, the capsules break and healing agent is pulled into the crack by capillary action.

The encapsulation strategy is capable of increasing the lifespan of chemical or biological healing agents and controlling their releases into the matrix. There is evidence suggesting that the capsule-based approach is versatile and the quality of repair is satisfactory, which is generally measured by recovery of mechanical and durability properties. However, the main challenge with the capsule-based approach is its repeatability over the long term. Concrete structures are subjected to multiple damage cycles throughout their service life and therefore a capsule-based system is expected to offer multiple instances of quality healing. Microcapsules can encapsulate limited amounts of repair agent and therefore most of the healing agent is exhausted under a single loading cycle and hence repeated healing over the long term is questionable. However, recent research efforts focused on smart release of healing agents [including those by Dong et al. (2015)]. Therefore, although not very established at this point of time, capsules may be designed so that multiple healing cycles can be achieved. Thao (2011) and Van Tittelboom et al. (2011b) evaluated the effectiveness of healing under multiple loading cycles by incorporating healing agent in tubes; they found that recovery of stiffness decreases under multiple cycles of loading. Therefore, future research on repeatability of capsule based systems is needed. Fig. 1 explains healing by the microcapsule-based approach when a crack ruptures the capsule (s) in its path. Depending on the encapsulation technique used, capsules are either strategically placed at predicted locations of failure (e.g., tubular glass capsules) or are dispersed throughout the matrix as uniformly as possible (e.g., microcapsules).

Capsule-based healing can be broadly categorized into healing induced by (1) bacterial precipitation and (2) encapsulated chemical healing agents. Studies on different properties of these healing mechanisms are discussed in the following sections.

## Comparison of Different Aspects of Capsule-Based Self-Healing

### Healing by Encapsulated Chemical Agent

#### Materials for Capsules

A variety of encapsulating materials, such as glass tubes, ceramic tubes, lightweight aggregates, and polymers have been used in developing self-healing action in concrete. Polymeric microcapsules are very frequently used and they are prepared by an oil-in-water dispersion mechanism of the polymer material, based mainly on the miniemulsion polymerization technique (Asua 2002). Urea and formaldehyde are made to react in the liquid phase, which eventually becomes cross-linked to form the urea formaldehyde (UF) capsule shell wall. The prepolymer that is formed by reaction in

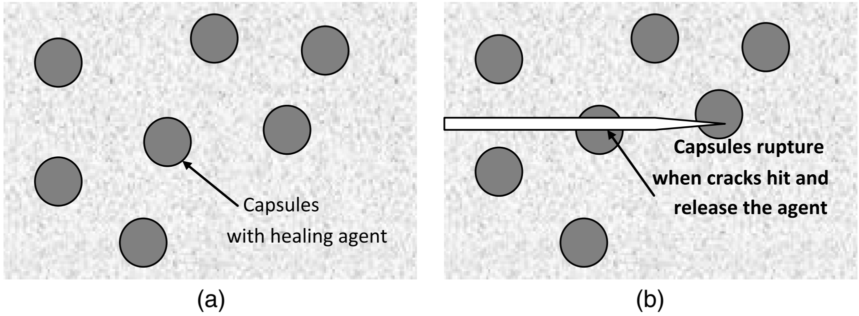

**Fig. 1.** Schematic of capsule-based self-healing approach

the water phase can be deposited to give a rough texture to the microcapsules. This can help to improve bonding with the cementitous matrix. Such capsules with healing agent have been used by Kessler et al. (2003), Brown et al. (2005), and Feng et al. (2008).

Blaiszik et al. (2009) reported a method using a sonification technique and hydrophobic solution for stabilization of a dicyclopentadiene (DCPD) healing agent for synthesizing UF microcapsules. Capsules with diameters of 220 nm and shell wall thickness of 77 nm were produced successfully with a more-uniform shell wall. However, the literature mentions several problems with nanosized capsule debris accumulating in the host matrix and these can even initiate cracking.

Apart from UF, melamine-based and polyurethane (PU) capsule materials have also been used in self-healing applications. Pelletier et al. (2011) and Liu et al. (2009b) prepared melamine-urea-formaldehyde (MUF) capsules that were found to have thermal stability up to 300°C. In fact, MUF microcapsules have been reported to demonstrate properties more superior than those made only from UF; synthesis of the former is also easier (Aïssa et al. 2012).

Thao et al. (2009) and Thao (2011) conducted comparative studies between Perspex cast acrylic tubes and glass tubes; they found that glass tubes were more suitable because of their inertness to the contained polymer. The brittle nature of the tubes also implies that it can be ruptured more easily. Optimal dimension for internal and external walls was found to be 4 and 6 mm respectively.

Van Tittelboom and De Belie (2010) and Van Tittelboom et al. 2011b) modified that system design by using glass and ceramic cylindrical capsules of different diameters. Capsules were placed next to one another so that when they ruptured together, the healing agents could mix easily. Different healing agents such as epoxy resins, polyurethane, and methyl methacrylate (MMA) were tested; the best result was obtained from the MMA–polyurethane combination (Van Tittelboom et al. 2011b). Glass capsules of 3 mm diameter performed best in terms of recovery of strength, while ceramic capsules performed best in terms of recovery of water permeability. In this research, glass capsules were located at predicted locations of cracks. For most building components, prediction of crack location may not always be possible. It may be better to randomly but uniformly distribute the capsules in pairs in all those areas that are vulnerable to cracking.

## Materials for Healing Agents

Huang et al. (2011) and Pelletier et al. (2011) used spherical capsules to encapsulate a sodium silicate solution. Rupture of capsules released the solution into the matrix and the reaction took place with calcium hydroxide to form calcium silicate hydrate (C-S-H) that healed the concrete crack. Li et al. (2013) encapsulated epoxy in polystyrene-divinylbenzene (St-DVB) microcapsules. An amine-based hardener was separately dispersed into the matrix. Once it properly cured, the epoxy that was released upon rupture of the capsules reacted with the hardener and healed the cracks.

Thao et al. (2009) and Thao (2011) used isocyanate prepolymer encapsulated in hollow cylindrical glass tubes. Selection of healing agent, encapsulation material, and protection concept in concrete were studied. The epoxy polymer that was chosen had low viscosity between 250 and 500 mPa s, and so it could flow smoothly into cracks and provide effective healing.

Cailleux and Pollet (2009) used microcapsules to hold bisphenol-F epoxy resin and embedded these microcapsules in the concrete, in which a hardener was separately dispersed. Healing takes place through a polymerization reaction of the epoxy resin with the hardener. They also encapsulated tung oil or calcium hydroxide with spherical microcapsules made of a gelatin shell. However, a problem faced was the premature rupture of capsules during mixing. Improvement in capsule design, in terms of material choice and thickness, was required to prevent them from being destroyed during mixing. Similar principle was applied by Dry (1994a, 1999), using cylindrical wax-coated PP capsules containing an MMA monomer as the healing agent. In particular, Dry (1999) used glass capsules with cyanoacrylate (CA). When these glass tubes broke under load, the CA was released into the medium and healed the cracks.

In other research efforts (e.g., Yang et al. 2010, 2011), microcapsules made of silica gel with oil core were used. The oil core phase consisted of MMA monomer as the healing agent and triethylborane (TEB), which acted as catalyst to heal microcracks.

## Capsule Geometry and Special Design for Mixture

Several researchers, including Joseph et al. (2007, 2010), Van Tittelboom and De Belie (2010), and Sun et al. (2011), noted that insufficient capillary force of the crack and gravitational pressure of the fluid mass will cause lower than expected amount of healing agent being released into the matrix. Cylindrical capsules in many occasions suffer from resistive capillary force and negative pressure at both ends. An illustration of forces acting on tubular shaped capsules is shown in Fig. 2. This problem was to some extent ameliorated by Li et al. (1998) using high quantities of polyethylene (PE) fibers to control crack width and thereby increasing the capillary attraction of cracks to release the healing agent. De Rooij et al. (2009) and Liu et al. (2009a) proposed the usage of coated hollow plant fibers as encapsulation to reduce the negative pressure at the sealed ends of glass capillary tubes. Fiber bundles delaminated when encountering propagating cracks, thus releasing the healing agent at the damaged sites.

In a multicapsule system, two or more different types of capsules store different healing agents. Healing is triggered by rupture of both types of capsules, causing them to react with each other.

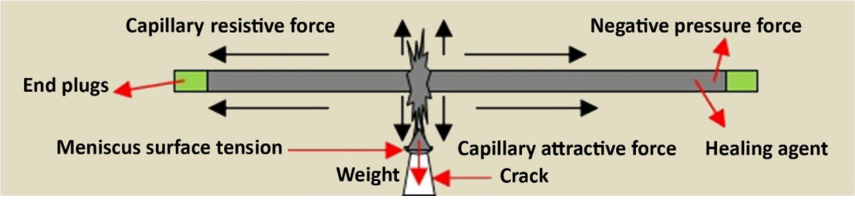

**Fig. 2.** Schematic illustration of forces acting on a capillary tube with healing agent

Mihashi et al. (2001) used spherical capsules with a urea formaldehyde formalin (UFF) shell containing two-component epoxy. However, healing was ineffective, because inadequate mixing prevented hardening of two-component epoxy. This process was improved by Feng et al. (2008), who modified the epoxy resin with a dilutant chemical to adjust the viscosity to yield better mixing. Although the reaction could occur at room temperature, the full benefit could only be obtained upon thermal curing at around 120°C.

Kaltzakorta and Erkizia (2011) encapsulated a two-component epoxy system by silica microcapsules. This direction of research is only in the initial phase and therefore the concept of mixing this type of capsule in the cement paste has not been rigorously proven.

## *Healing by Encapsulated Bacteria*

### Encapsulation in Polymer and Special Mineral Compound

Wang et al. (2014b) encapsulated spores of *Bacillus sphaericus* in a melamine-based microcapsule system that also contained an inert material for protection of the spores. Bacteria concentration of about $10^9$ cells/g of dry microcapsule was added. Crack healing efficiency measured in terms of the ratio of healed crack to initial crack area showed a maximum healing rate of 80% (under wet-dry curing); maximum healed crack size recorded was 970 $\mu$m (under water curing).

In an earlier study, Wang et al. (2012a) used diatomaceous earth (DE) as a carrier for *Bacillus sphaericus*. This species was made to react with hydrolyzed urea provided in the cementitous matrix to precipitate calcium carbonate. Cracks of width between 0.15 and 0.17 mm were found to be partially or completely healed. However, DE tends to absorb high quantity of water and dry up the mortar. Another problem is the formation of small amount of excess urea or calcium nitrate crystals that may affect concrete strength.

### Encapsulation in Special Cement Aggregates

Jonkers (2011) and Wiktor and Jonkers (2011a, b) used expanded clay aggregates to embed a two-component healing agent comprising of bacteria and calcium lactate. Porous clay aggregates (lightweight aggregates (LWA) act as an internal source of moisture that is necessary to support bacterial precipitation action. However, the efficiency depends on a number of parameters, such as the amount of water in the aggregate, aggregate spacing, and aggregate pore structure (Reynolds et al. 2009). There is a limitation to the use of expanded clay encapsulation. When natural aggregates are replaced with clay LWA, there is substantial reduction in the mechanical strength of concrete. Aggregates form the bulk of concrete and the compressive strength of concrete is substantially determined by their toughness. As much as 50% strength reduction was observed after 28 days (Jonkers 2011), which is not desirable for structural applications. When the clay particles ruptured, calcium precipitation by microbial action occurred as a result of the bacteria spores coming in contact with air. After 100 days of immersion in tap water, maximum crack width that could be healed completely was about 0.46 mm. Activity up to 6 months has been observed so far.

### Encapsulation in Special Additives in the Cement Matrix

Wang et al. (2014a) used hydrogel-microencapsulated bacteria spores and bioreagents (nutrients for bacteria, urea, and calcium nitrate in this case) in their study. The main advantage of hydrogel is its capability to absorb and retain moisture over a long period of time. The encapsulated spores could grow into active cells and precipitate calcium carbonate by urea decomposition when there were sufficient water and nutrients in the hydrogels. Maximum crack width healed was around 0.5 mm and a maximum decrease in water permeability of about 68% was observed.

Table 1 summarizes the findings from key literature in which carbonate-precipitating bacteria is used as a self-healing agent. However, not all the species have been used as healing agent inside capsules.

Table 2 summarizes the advantages and disadvantages of each of discussed self-healing approach. Different healing agents and encapsulation materials, as well as casting and mixing techniques used in developing self-healing concrete, are compared in Table 3.

## Efficiency of Various Self-Healing Materials to Yield Similar Output

Regardless of the method used, the objective of self-healing is to achieve as high as possible healing output, which may be measured by crack width sealed and recovery of original concrete properties. Before selection of any self-healing method for actual site application, it is, therefore, essential to enumerate efficiency of different methods to offer similar output.

As already discussed, the microcapsule technique has been used to encapsulate different healing agents including chemicals, polymers, and bacteria spores. Pelletier et al. (2011) encapsulated sodium silicate solution in polyurethane (PU) microcapsules (size 40–800 $\mu$m) which were introduced into cement mortar. Recovery of flexural strength was about 20–26%, while for control samples only about 9–10% of strength was recovered. Li et al. (2013) encapsulated epoxy in polystyrene-divenyl benzene (St-DVB) capsules, which were added by 1 and 2% by mass of cement. Specimens were damaged to 30, 60, and 100% of ultimate load. It was observed that under standard curing (90% RH, 20 ± 2°C), normalized flexural strength recovery was highest (about 1.8) when 1% capsules were added and damage level was 30%. However, in case of water curing, 2% microcapsule addition at 60% damage produced best result (normalized strength recovery of 1.4). No data pertaining to maximum crack width sealed were available although it may be concluded from the studies that healing efficiency was dependent on balance between extent of damage (or crack width created) and volume of microcapsules added. Lower strength recovery in case of water curing may be the result of deteriorated hardening and

**Table 1.** Review of Bacterial Species and Their Functions in Self-Healing (Data from Gupta et al. 2013)

| Species of microorganism | Mechanism | Major findings | References |
|---|---|---|---|
| *Bacillus pasteurii*, *Pseudomonas aeruginosa* | Urease metabolism | More effective as crack sealant but less effective in increasing compressive strength<br>Higher efficiency in shallow cracks due to higher availability of oxygen | Ramachandran et al. (2001) |
| *Bacillus pasteurii* | Polyurethane (PU) immobilized, urease metabolism | Immobilization technique retains high cellular metabolism and protects the cells against harsh environment<br>Higher bacteria cell concentration per crack helps to regain higher early (7 days) compressive strength | Bang et al. (2001) |
| *Bacillus sphaericus* | Ureolytic decomposition of calcium nitrate | Concrete permeability reduced<br>Crack widths between 150 and 170 $\mu$m completely healed | Wang et al. (2012a) |
| *Bacillus pseudofirmus* and *Bacillus cohnii* | Metabolism of calcium lactate | 20–80 $\mu$m calcium crystals were seen on crack surfaces<br>Performance limited to young concrete | Jonkers et al. (2010); Jonkers (2011) |
| *Bacillus cohnii, Bacillus halodurans*, and *Bacillus pseudofirmus* | Two component system | Substrate and microbe added into the concrete<br>Can seal cracks up to 100 $\mu$m | Jonkers and Schlangen (2009) |
| *Bacillus sphaericus* | Ureolytic decomposition of calcium nitrate | Maximum crack sealing of 500 $\mu$m under wet-dry cycles<br>Permeability decreases by 68% for specimens containing hydrogel encapsulating both bacteria and nutrients together | Wang et al. (2014a) |
| *Bacillus sphaericus* | Ureolytic decomposition of calcium nitrate | Alginate (type of hydrogel) protected about 90% of the spores<br>Bacterial activity was observed only for encapsulated samples at crack face measured by oxygen consumption | Wang et al. (2015) |

**Table 2.** Summary of Advantages and Disadvantages of Self-Healing Approaches

| Approach | Strategy | Advantages | Challenges/disadvantages |
|---|---|---|---|
| Autogenous healing | Hydration of unhydrated cement particles | High recovery of mechanical properties possible<br>Healing possible under different exposure conditions | Using high amount of cement is not sustainable<br>Performance depends on age of concrete and age of cracking |
| | Dissolution and carbonation of calcium hydroxide | | Significant healing can be achieved only if there is tight control of crack width, below 150 $\mu$m and preferably below 50 $\mu$m (Yang et al. 2009; Van Tittelboom et al. 2016). |
| | Supplementary cementitous material (SCM) and expansive agent | Healing late in the service lifespan of concrete is possible due to slow hydration of SCMs<br>Satisfactory healing of cracks<br>Healing agent compatible with cement matrix | Continuous exposure to water is needed<br>Expansive agents may cause microcracking in the matrix<br>Healing under multiple loading may not be possible |
| Vascular self-healing | Vascular approach | Large amount of healing agent can be supplied and so macro-cracks can be healed<br>Potential for higher recovery of mechanical strength and durability | Difficult to cast network of tubes on bigger scale.<br>Introduction of supply tubes may weaken the concrete structure<br>Strategic placement of glass capsules is needed at zones of cracking |
| Capsule-based system | Encapsulation of chemicals (glass tube and microcapsules) | Respond to multiple cracking at the same time<br>Satisfactory recovery of mechanical and durability property | Introduction of many glass capsules can weaken concrete structure<br>Recycling rate of concrete with chemical agents may be low, and disposal may not be environmentally friendly<br>Inadequate data on repeatability for multiple loading cycles |
| | Encapsulation of bacteria (microcapsules) | Environmentally friendly mechanism | Repeatability under multiple damage cycles is unlikely<br>Effectiveness is highly dependent on availability of moisture |
| | | Precipitate compatible with matrix | High cost of production of axenic spores for effective self-healing |

**Table 3.** Summary of Some Capsule Material and Healing Agent Used in Developing Self-Healing Concrete

| Approach | Healing agent | Number of components | | Viscosity (mPas) | Capsule material | Capsule dimension | | | Mixing/casting technique | References |
|---|---|---|---|---|---|---|---|---|---|---|
| | | Single | >1 | | | Diameter ($\mu$m) | Wall thickness ($\mu$m) | Length (mm) | | |
| Encapsulation of chemical agents | Cyanoacrylate (CA) | 1 | — | <10 | Glass | 800<br>1,500<br>3,000 | X<br>X<br>X | 75<br>75<br>100 | Tubes filled with CA with ends plugged with wax, were placed in single layer or double layer in concrete | Joseph et al. (2010) |
| | CA | — | 2 | <10 | Glass | 2,000–3,000 | 100 | 20–80 | Glass tubes filled with healing agent were placed in the concrete before casting | Van Tittelboom and De Belie (2010) |
| | Epoxy | — | 2 | 150<br>80<br>360 | Glass | 2,000–3,000 | 100 | 20–80 | | |
| | Epoxy | 1 | — | 250–500 | Glass<br><br><br>Perspex | 5,000<br>6,000<br>7,000<br>X | X | 250<br>250<br>X<br>X | Glass tubes were placed at depth of 10 mm from bottom of concrete specimen | Thao et al. (2009) |
| | Epoxy | — | 2 | X | UFF | 20–70 | X | NA | Spherical UFF capsules containing two-component epoxy were mixed in mortar | Mihashi et al. (2001) |
| | Acrylic resin | — | — | X | Gelatin | 125–297 | X | NA | | |
| | Epoxy | — | 2 | 200 | UF | 120 | 4 | NA | Spherical microcapsules containing two-component epoxy and dilutant chemical mixed with concrete | Feng et al. (2008) |
| | Sodium silicate solution | 1 | — | X | PU | 40–800 | X | NA | Capsules after encapsulation were added to water and mixed with mortar | Pelletier et al. (2011) |
| | MMA with TEB as catalyst | — | 2 | 1 | Silica gel | 4.15 | X | NA | Microcapsules with MMA oil core were added to cement and mixed with capsules containing TEB | Yang et al. (2011) |
| | Epoxy | 1 | — | X | Gelatin | 50 | X | NA | Healing agent encapsulated in spherical gelatin capsules with and mixed with mortar | Cailleux and Pollet (2009) |
| | Tung oil | 1 | — | X | Gelatin | 50 | X | NA | | |
| | Calcium hydroxide | 1 | — | X | — | 50 | X | NA | | |
| | Polyurethane (PU) | —<br>— | 2<br>2 | 600 | Glass<br>Ceramics | 2,200–3,350 | 100 | 20–80<br>15–50 | Tubes containing prepolymer and accelerator were embedded in concrete samples | Van Tittelboom et al. (2011b) |
| | | — | 2 | X | Glass | 3,350 | 350 | 50 | Tubes containing PU manually placed. Ends sealed by MMA | Van Tittelboom et al. (2016) |
| Encapsulation of bacteria (producing calcium carbonate as the healing agent) | Bacteria and calcium lactate | — | 2 | NA | Expanded clay | 1,000–4,000 | NA | NA | Clay particles loaded with lactate and bacteria were oven-dried at 40°C and added to concrete | Jonkers (2011) |
| | Bacteria | —<br>— | 2<br>2 | NA<br>NA | PU in glass<br>Silica gel in glass | 2,200–3,350 | 100 | 20–80 | PU with bacteria and silica gel with bacteria injected in glass tubes. Tubes were glued to 10 mm mortar layer and cast | Wang et al. (2012b) |
| | Bacteria | 1 | — | NA | Melamine | 5 | X | NA | Capsule emulsion was mixed with water and added to mortar | Wang et al. (2014b) |
| | Bacteria | 1<br>— | —<br>2 | NA<br>NA | Hydrogel | NA<br>NA | NA<br>NA | NA<br>NA | Spores (with and without nutrients) and hydrogel were mixed with mortar | Wang et al. (2014a) |
| | Bacteria | 1 | — | NA | Diatomaceous earth (DE) | X | NA | NA | DE and bacteria were mixed with cement, sand, and nutrient solution | Wang et al. (2012a) |

Note: X = not reported; NA = not applicable.

setting of epoxy in cracks due to presence of liquid water. In contrast, water is the most essential component when bacteria spores are encapsulated. Wang et al. (2014b) observed up to 80% healing ratio (measured by closure of crack area) and maximum crack width closure close to 0.6 mm under a wet-dry cycle of curing when *Bacillus sphaericus* spores were encapsulated in melamine microcapsules. The co-efficient of permeability in all bacteria healed specimen was in the range of $6.4\times10^{-8}$ to $1.6\times10^{-7}$ m/s, meaning maximum improvement of an order of magnitude over nonbacterial samples. Healing ratio was improved (up to 90%) when bacteria spores were encapsulated in hydrogel although maximum crack width closed was still about 0.5 mm (Wang et al. 2014a).

Tubular encapsulation is one of the widely studied self-healing techniques used for cementitous materials. Maes et al. (2014) studied improvement in durability by incorporation of encapsulated PU in glass tubes for self-healing. Durability was measured by reduction in chloride penetration upon self-healing after crack widths of 100 and 300 $\mu$m were introduced. It was observed that autonomous healing by PU is able to seal crack widths of 100 and 300 $\mu$m for chloride penetration in 67 and 33% of the cases. However, large uncertainties in test results were observed, which as attributed to insufficient release of healing agent from glass capsules. Previously, it has been established that healing by encapsulation of PU in glass and ceramic tubes is able to reduce water permeability by a factor of up to $10^3$ and $10^4$, respectively, (Van Tittelboom et al. 2011a) when a crack width between 200 and 300 $\mu$m is introduced. In addition, average recovery in strength and stiffness over 50% has been recorded. When bacteria spores were encapsulated in glass tubes by immobilization in PU, strength recovery between 50 and 80% was observed, although it was primarily due to PU rather than calcium carbonate precipitated by bacteria (Wang et al. 2012b). It was observed that water permeability could be reduced by factor of about $10^5$ compared to reference samples although it was only slightly lower than samples with polyurethane (without spores).

Therefore, the conclusion is that when using microcapsules, high healing output is largely dependent on optimization of dosage of capsules and control of crack width. When glass or ceramic capsules are used, they are already placed at strategic locations and therefore, to achieve high healing ratio it must be made sure that the tubes rupture upon cracking and healing agents flow out efficiently to the crack site.

In general, ensuring successful self-healing action by encapsulation requires optimizing, or at least, balancing different properties of the healing agent, capsule, and system design. Key properties can be categorized under the following eight aspects:

1. Robustness during mixing;
2. Probability of cracks encountering the capsules;
3. Curing time of healing agents and curing conditions;
4. Effect of empty capsules on concrete strength;
5. Controllability of release of healing agent;
6. Stability of healing agent;
7. Sealing ability; recovery of durability and strength of concrete matrix; and
8. Repeatability of self-healing action.

## Eight Effectiveness Factors to Evaluate Self-Healing System

### Robustness during Mixing Process

Glass capsules have been widely used to encapsulate healing agents. The advantage of using glass capsules lies in their brittleness, because they can rupture easily when a crack appears. However, the same property hinders their survival when the concrete is being mixed unless some form of protection is in place, like cement mortar or metallic wire (Thao 2011). Furthermore, addition of capsules must not adversely impact concrete workability. Thao (2011) tested three protection methods for tubular capsules: mesh, spiral wire, and spiral wire coated with mortar. Mortar thickness of 3.5 and 6.5 mm were used. Experimental results showed that protection by spiral wire coated with mortar with thickness about 3.5 mm was the most effective.

Hilloulin et al. (2015) explored development of polymeric capsules that can survive the mixing process without any protection. Polymeric materials for the capsules were selected so that they have a low glass transition temperature and were brittle at room temperature. Three types of polymeric materials were selected: polylactic acid (PLA), polystyrenene (PS), and polymethyl methacrylate/*n*-butyl methacrylate [P (MMA/*n*-BMA)]. It was found that capsules preheated and treated with boiling water had higher survival rate due to flexibility caused by heating above their transition temperature.

Van Tittelboom et al. (2015) measured the healing efficiency of cracks using the water permeability test, where short glass tubes with ceramic capsules were used with three different protection mechanisms: attaching to reinforcement, covering the tubes/capsules with one layer of mortar at the bottom of the mold, and embedding them in cement paste bar. It was observed that capsules attached to reinforcement bars survived the concrete mixing process without any other protection means; however, good workmanship is required to cast beams in different layers to implement this scheme. Capsules surrounded by the mortar layer performed best and since they were placed near the soffit, they were efficient in healing only surface cracks.

Wang et al. (2014b) used a light microscopy method to visualize the survival of melamine-based microcapsules (around 5 $\mu$m in dimension) that encapsulated bacteria spores. Comparing images taken before and after mixing, the authors concluded that microcapsules were not broken after mixing.

However, when bacteria spores are used, survival of bacteria is more important than survival of the capsules. Thus, even if a small number of capsules break during mixing, it may not be detrimental to the self-healing capability, as long as the bacteria spores can continue to precipitate calcium carbonate.

Finally, the wall thickness of the capsules is an important quality that protects the capsules during mixing. Premature rupture may occur if very thin walled capsules are used; on the other hand, if wall is too thick, it can prevent or delay the release of healing agent even when the capsule ruptures. This means that capsule wall thickness need to be adjusted accurately for efficient self-healing. Alternatively, bundling can increase survival rate of capsules. Dry (1996) used water soluble adhesive to bundle glass pipettes containing a healing agent so that capsules did not rupture during mixing. These capsules were dispersed in the concrete after the glue dissolved in water during mixing.

### Probability of a Crack Encountering the Capsules

When glass tubular capsules are used in the matrix, they create a weak plane that draws the cracks towards them. Compared to cylindrical microcapsules, the sites of cracking cannot be predicted with certainty for spherical and cylindrical microcapsules. The probability of cracks hitting the capsules increases with the number of capsules, but this reduces the strength of the concrete. Therefore, there should be an optimum balance between number of capsules and healing effectiveness.

Zemskov et al. (2011) developed analytical models to predict the probability of a spherical capsule encountering a crack. These models allow for the estimation of crack width, intercapsular distance, and capsule size for effective self-healing. They presented two models: random placement of spherical capsules in layers in the matrix, and entirely random dispersion of spherical capsules throughout the matrix.

In the first model, the probability of crack hitting a capsule of the first layer is a function of three parameters: crack depth, average capsule section radius, and healing agent content ratio. The second model could give more-accurate probability prediction of crack depths less than ($V^{1/3} - R$), where $V$ is the volume of the representative cube and $R$ is the capsule section radius. On the contrary, the first model is more flexible, because the probability function can be applied to any number of layers and cracks of any depth. However, it does not take into account crack widths and number of cracks formed, which may affect the probability of encountering a capsule. For example, if a large crack can be randomly dispersed into smaller cracks due to strain hardening, the chance of hitting randomly placed capsules (or also layered capsules) is higher.

A similar approach was used to develop a three-dimensional model to evaluate the necessary dosage of capsules, assuming penny-shaped cracks were randomly distributed throughout the matrix (Bejan et al. 2006). The optimization process accounted for the shape of the capsule, size of the capsule, and healing agent released from the capsule. Both spherically and cylindrically shaped capsules were considered, and the probabilities and capsule dosage were expressed in terms of capsule dimension for each shape.

Lv and Chen (2013) found that as the ratio of length of crack to radius of capsule increases, the probability of hitting also increases when aspect ratio (ratio of height to basic radius of cylindrical capsules, denoted by $\tau$) of capsules is fixed. The highest probability of crack hitting capsule was found for the lowest aspect ratio studied ($\tau = 0.5$), which means that reducing capsule size and increasing the number of capsules enhance the chance of hit. Under the same conditions, cylindrical capsules registered higher hit probability than spherical ones, due to higher surface-to-volume ratio. However, that study assumed that whenever a capsule ruptures, it completely heals a crack. The effect of crack width was also ignored.

Finally, while estimating the probability of a crack hitting the capsules, one needs to know that having too many capsules will weaken the material mechanical properties (Privman et al. 2007).

### Curing Time of Healing Agents and Curing Conditions

A good self-healing system must be able to respond to damage quickly, which implies that the encapsulated healing agent must be released as quickly as possible when required. Activation of encapsulated bacteria spores typically takes a few days to occur after it is exposed to moisture because the spores need time to grow. It was found that complete crack closure on the surface occurred after 2 weeks of water immersion (Jonkers 2011). It was also demonstrated by oxygen concentration measurement that bacteria and calcium lactate encapsulated in the matrix showed rapid oxygen consumption once the specimens were submerged in water. In fact, for rapid initiation of crack healing, bacteria and calcium lactate should be encapsulated together. Moreover, the efficiency of bacterial self-healing depends on the curing condition to which specimens are subjected after the introduction of cracks. For example, Jonkers et al. (2011) reported that best healing performance was achieved under water curing condition for 2 weeks when bacteria were encapsulated in LWA. Moreover, lightweight clay aggregates, if prewetted, can promote internal curing of concrete by releasing absorbed water over a period of time. Wang et al. (2014b) reported similar findings that specimen subjected to a 16 h wet-dry cycle for 3 weeks showed highest reduction in cracked area. This again proved that the presence of water is imperative for bacterial self-healing.

While using epoxy as healing agent, it must be noted that correct mix ratio is important for curing. Premature hardening, or delayed hardening, may take place when components are not properly mixed. Thao (2011) encapsulated one-component epoxy inside cylindrical glass capsules that cured in contact with air when released in the matrix. Due to lower reactivity of the agent, no hardening occurred before rupture of capsules. Thao (2011) observed that complete curing of the epoxy took place about 7 days after being released from the capsules. Crack width of 0.3 mm was healed, with 99% recovery of stiffness (in the case of concrete slabs). Furthermore, when concrete members are exposed to sunlight, the epoxy may cure faster because of polymerization induced by photochemical reaction. Although it is a low heat process, faster curing may take place by enhanced degree of cross-linking between constituent oligomers of epoxy.

Some research studies also reported application of heat to cure chemical healing agents. Dry (1994a, 2000) embedded porous cylindrical PP capsules encapsulating MMA and coated with wax in concrete beams. Upon crack appearance, the concrete beams need to be heated to melt the wax and release MMA through pores of PP capsules. MMA inside the cracks cured due to heating. Nishiwaki et al. (2006) also used heating (at 60°C) to reduce curing time of epoxy resin to only 100 min.

Curing time of healing agents also depend on their viscosity. MMA has been used as self-healing agent in a few studies (Dry and McMillan 1996; Dry et al. 2003; Yang et al. 2011). Due to its very low viscosity and a comparatively long curing time (longer than 30 min), MMA may be absorbed by the matrix and thus leave a partially healed crack behind. Dry and McMillan (1996) noted that MMA may even leak out of the damaged site, thus resulting in incomplete healing. Hence, modification of such low-viscosity healing agents is often required to make it suitable for crack filling.

### Effect of Emptied Capsules on Concrete Strength

Addition of healing agents and/or capsules can decrease the mechanical strength of concrete specimen, because many polymer/chemical materials create weak links in the concrete matrix, although their performance as capsule material is satisfactory. Also, after the healing agent is discharged, the emptied capsules act as voids inside the matrix. Therefore, higher dosage can actually lead to higher reduction in strength due to creation of more voids. For example, Wang et al. (2014b) used melamine-based microcapsules of 5 $\mu$m in dimension at different dosages (ranging from 1 to 5%) and recorded losses in compressive strength with increases in capsule dosage.

Other than dosage, capsule materials and dimension may influence concrete strength too. Pelletier et al. (2011) used spherical PU capsules whose diameters varied from 40 to 800 $\mu$m to encapsulate a sodium silicate solution. About 2% microcapsules were mixed into the matrix. Although capsules of dimensions bigger than those used in the study by Wang et al. (2014b) with similar dosage were used, no significant drop in compressive strength was recorded. Feng et al. (2008) used UF spherical microcapsules filled with epoxy. The diameter was 120 $\mu$m and wall thickness was 4 $\mu$m; no loss in mechanical strength was found.

Bigger encapsulating units, such as glass tubes, tend to introduce weaker planes in concrete and reduce the mechanical properties. This happens because of the weaker bond strength between the capsule and mortar and the lower tensile strength of capsules.

Hilloulin et al. (2015) showed that there is potential to replace glass capsules by polymeric capsules without affecting concrete mechanical strength. The experiment involved the use of three types of capsules [PLA, PS, and P(MMA/*n*-BMA)]. All the three capsule materials have higher tensile strength than concrete. Highest bond strength was found for PS capsules—around 3.5 MPa higher than average bond strength of glass, which varies between 0.2 and 1 MPa (Hilloulin et al. 2015).

### Release Efficiency of Healing Agent from Capsules

Once the capsules are ruptured, the healing agent is expected to be released and deployed into the matrix quickly. In this regard, spherical capsules could be better than cylindrical ones, because the former has a more-uniform shape and the suction effects at closed ends of cylindrical capsules makes release of healing agent difficult for these capsules. Experiments carried out by Jung (1997) highlighted that spherical capsules allow lower transport distance of the healing material in cracks compared to elongated capsules. However, this enhanced release of healing agents from spherical capsules could be a problem due to rapid exhaustion of healing material (Mihashi et al. 2001). Feng et al. (2008) reported that using epoxy as healing material inside spherical capsules could heal only a few cracks due to quick exhaustion of materials. On the contrary, cylindrical capsules showed significant improvement in recovery and better mechanical properties, such as strength and durability.

Van Tittelboom et al. (2015) studied the release efficiency of healing agent with two types of tube dimensions. Long continuous tubes were not able to release healing agent after breakage because resistance force inside the 3-mm-thick long capsules were high. In contrast, shorter capsules with a length 60 mm (and 3 mm thickness) were found to be more efficient in terms of release of agent and crack sealing.

Dong et al. (2015) worked on development of smart-release chemical microcapsules targeted at healing concrete degradations, especially corrosion of reinforcement bars triggered by reduced alkalinity of concrete matrix. Corrosion initiates cracking and hence, such development may indirectly prevent cracking by realkalization, for example, by decreasing chloride or acidic concentration in the pore solution. PS and sodium monofluorophosphate ($Na_2PO_3F$, MFP) were used to make the microcapsules. Release of the healing agent was influenced by capsule shell thickness and the pH value of the matrix. Under normal alkaline condition, calcium ion reacts with phosphate ion to form precipitate that seals the capsule pores and prevents further release of materials; the healing agent is released by diffusion once low-pH conditions result in the formation of pores in the capsule. However, this is still at preliminary stage and the effectiveness of corrosion and crack prevention in concrete has not yet been tested.

Mookhoek (2010) and Mookhoek et al. (2009) documented that for same capsule volume fraction, larger amounts of healing agent could be delivered to a certain cracked location by cylindrical capsules, and thus healing of larger crack volume is more effective by cylindrical capsules. It could be inferred from a simulation study by the representative volume element (RVE) approach that the average volume released per area is higher for cylindrical capsules for a fixed aspect ratio compared to spherical capsules of same radius. Orientation of cylindrical capsules also influences the release of the healing agent. The release efficiency will be the highest when cylindrical capsules are oriented perpendicular to the crack plane. This is due to highest probability of rupture (Mookhoek 2010).

Nevertheless, controllability of release would also be influenced by viscosity of encapsulated healing agent. Highly viscous healing agents may not be fully deployed to the crack site from the capsules, whereas if the viscosity is very low, the healing agent may leak out through the crack it aims to heal. Although CA has relatively low viscosity, it is often used as a self-healing agent to seal macroscopic cracks. Due to its low viscosity, it can flow into nearby microcracks as well (Joseph et al. 2010). Dry (1994b) observed that suitable healing agents should have viscosity between 100 and 500 centipoise (cps) and the short curing time (only a few seconds) of CA also resulted in fast recovery in mechanical properties after crack formation.

Epoxy resin is also widely used as healing agent but it is known to be highly viscous. Since resins of different viscosity are available commercially, epoxy resin may be modified by adding diluting chemicals to it (Feng et al. 2008). For example, Van Tittelboom et al. (2011a) mixed poly-methyl methacrylate (PMMA) with epoxy to increase the viscosity by thickening the healing agent and retaining it inside the crack site.

### Stability of Healing Agents

Healing agents used must be stable over time to heal cracks at later stages of a building's life span. Most of the single-component healing agents react with moisture or air. Therefore, if there are any trapped air bubbles inside the capsule or moisture diffuses through the capsule wall, the healing agent may harden and stability is compromised (Li et al. 1998; Van Tittelboom and De Belie 2010). Heating as a result of exposure to sunlight may also result in premature hardening of healing agent and compromise the recovery of durability and mechanical properties.

In general, a two-component healing agent is more stable than a single-component one because it reduces the chance of premature activation of the healing agent. However, there are concerns regarding the fact that using separate capsules for a two-component healing system may cause the healing agents not to be released from both the capsules evenly, thus resulting in improper mixing. However, a well-designed system and careful selection of healing agent may eliminate the risks. The two-component healing agent used by Van Tittelboom et al. (2011b) had relatively low viscosity and the polymerization reaction between the components did not depend on the mix ratio of the components. Since an expansive reaction occurred, it acted as a driving force to further push healing agent out of the respective capsules.

For bio-based systems, stability of bacteria is directly linked to their survival rate. Dormant bacteria spores are more stable than live cells, because the former can withstand heat and mechanical stress over a long period of time. However, Jonkers (2011) noted that the lifetime of the unprotected spores was limited to only 2 months, and therefore, effective self-healing was observed only in young cement samples. There may be several reasons for this, including alkalinity of cement matrix, mixing of concrete, and hydration of cement. Therefore, expanded clay aggregates were used to encapsulate the bacteria spores and viability study conducted report no loss of activity up to 6 months.

### Sealing Ability; Recovery of Durability, and Strength of Concrete Matrix

Efficient self-healing in concrete would mean that the durability and mechanical strength are fully recovered or close to that of the original specimen. Recovery of mechanical and durability properties is high if cracks and pores are blocked effectively. In other words, the sealing ability of the healing agent influences the degree of recovery of original concrete properties.

Durability is often measured by water permeability and water absorption tests, while recovery of strength may be determined by mechanical tests including test for compressive and flexural strength and stiffness. Table 4 summarizes techniques used to test the recovery of mechanical and durability properties after self-healing has occurred.

## Recovery of Mechanical Strength

The strength of healing agent must be equal or higher than the host matrix for higher recovery of strength. Healing a crack with a healing agent that is weaker than the matrix may not be effective because the healed spot creates a weak zone and stress concentration, and cracks may start to propagate under further loading (White et al. 2001).

The literature reports tensile strength of some commonly used polymers; for example, cured CA is about 20 MPa (Joseph et al. 2010), PMMA is in the range of 50–75 MPa, and epoxy resins may range from 5 to 45 MPa (Thao et al. 2009; Kuang and Ou 2008). Generally, tensile strength of concrete is much lower than the tensile strength of these polymers. Even with a very low water–binder ratio for high-strength concrete that incorporates steel fibers, split tensile strength of concrete may reach between 10 and 12 MPa depending on the fiber volume added (Song and Hwang 2004).

One of the challenges with the capsule-based approach is the limited amount of healing agent that can be encapsulated, which may be insufficient to heal the cracks in some cases. This problem can be overcome by using agents that expand upon polymerization, thus enabling them to plug and seal bigger cracks with smaller encapsulated volume. Van Tittelboom et al. (2011b) used PU as a healing agent that healed cracks by expansive reaction. The reaction product did not only fill the cracks but acted as a driving force to push healing agents out of the capsules. However, expansive reactions may cause internal cracking if the expansive strain is higher than strain capacity of concrete [as observed by Sisomphon et al. (2011b)]. Although the encapsulation technique was not used, many microscopic cracks were observed when ettringite was used to heal cracks by expansive reaction.

Stiffness of the healing agent and its bonding with the concrete matrix is necessary for high recovery of mechanical strength. Although stiffer healing agents impart higher stiffness to the structure, movement is restricted due to their inherent brittleness. Elastic

**Table 4.** Review of Test Methods Used to Assess Healing and Recovery of Mechanical and Durability Properties after Healing

| Type | Test | Purpose | References | Limitations |
|---|---|---|---|---|
| Visualization and determination | Scanning electron microscopy | Visualization of crystal deposited for healing and release of healing agent in smart capsules | Cailleux and Pollet (2009), Huang et al. (2011), Wang et al. (2012a, b, 2014a, b), Dong et al. (2015) | Image accuracy and visualization is dependent on where the image is taken and the resolution chosen<br>It may not capture the uniformity of deposition |
| | Infrared analysis | Determination of precipitated products | Wiktor and Jonkers (2011b) | Presence of moisture in concrete may affect accuracy<br>Infrared is suitable only to see prominent components and thus minor depositions may not be discernable (Ramachandran and Beaudoin 2000) |
| | Environmental scanning electron microscopy | Visualization of rupture of capsules embedded | Li et al. (1998), Sisomphon et al. (2011a) | Very low pressure that has to be maintained may alter the microstructure of concrete by dehydrating it |
| | Optical microscopy with image analysis | Visualization of crystal deposition and healing rate | Wiktor and Jonkers (2011b), Wang et al. (2012a, 2014b) | Depends on the resolution of optical microscope, which may be limited by the thin section |
| Recovery of water and air tightness (durability features) | Water permeability (low pressure and high pressure) | Water permeability coefficient can be determined by flow of water through healed cracks | Jonkers (2011), Van Tittelboom et al. (2011b, 2015), Wang et al. (2014a, b) | Effectiveness is dependent on how the cracks were introduced |
| | Air permeability | Flow rate of air after healing has occurred measures the resistance against moisture/foreign substance penetration through healed cracks | Yang et al. (2011) | Very sensitive to composition of the specimen |
| | Chloride diffusion | Measurement of resistance against chloride penetration. Relevant and applicable for coastal structures | Termkhajornkit et al. (2009) | |
| Recovery of mechanical properties | Compression test and tensile test | Measure recovery in strength due to self-healing | Yang et al. (2011), Qian et al. (2009) | Strongly influenced by moisture content, size and curing of specimens |
| | Bending test (three-point and four-point) | | Pelletier et al. (2011), Van Tittelboom et al. (2011b, 2015, 2016) | Resonance frequency results are affected by size and geometrical effect of specimen (Waiching et al. 2015) |
| | Resonance frequency analysis | Measurement of recovery of stiffness | Abd-Elmoaty (2011) | |
| | Acoustic emission analysis | Signals from sensors that are attached to surface are captured and analyzed to detect capsule breakage and regain in energy | Van Tittelboom et al. (2012, 2015) | Sensitive to the quality of environmental noise and signal (Huang et al. 1998) |

behavior prevents bond loss with the matrix and enables the matrix to respond well to thermal movements and cyclical loadings. Dry et al. (2003) mixed foam materials and beads that are compressible with epoxy resin. The research concluded that both stiff (high modulus of elasticity) and flexible additives (low modulus of elasticity) transferred stress equally well across microcracks, which may be due to the fact that effective modulus depends on cross section and there is little room for movement and change in cross section in the case of very small cracks.

Van Tittelboom et al. (2016) studied two mechanisms of self-healing, including encapsulated polyurethane (PU) and directly added superabsorbent polymers on real scale beams (150 × 250 × 3,000 mm). The capsules were placed by a network of plastic wires that was connected with the walls of the mold at a depth of 10 mm. After introduction of cracks by means of four-point loading, the beams were allowed to heal for 7 weeks. It was observed that cracks up to 189 $\mu$m could be closed completely by the released PU upon rupture of capsules. Tomographic analysis indicated that, although PU could not form a continuous layer when sealing the cracks, the sealing was nonetheless sufficient to restrict the passage of foreign elements through the cracks. However, for some zones, crack healing by PU was not observed, which was attributed to crack propagating around the capsule without rupturing it. For SAP samples, more-complete crack filling was observed by means of further hydration and precipitation of calcium carbonate. Cracks with width up to 198 $\mu$m were almost completely healed, while partial healing occurred for cracks up to a width of around 600 $\mu$m. While healing is more uniform in samples containing SAPs, the biggest advantage of PU-based healing is its ability to heal without any moisture.

### Recovery of Durability

Van Tittelboom et al. (2011b) measured decrease in water permeability for cracks created with width between 200 and 300 $\mu$m. The self-healing system consisted of glass or ceramic tubes with two tubes positioned next to each other, one containing prepolymer and another containing accelerator and water. Results showed that when glass tubes were used, water permeability coefficient decreased substantially after healing; for ceramic tubes, the decrease was even higher. This difference may be due to higher release of healing agent from ceramic tubes, which can be explained by surface tension between the glass and ceramic. Yang et al. (2011) measured reduction in gas permeability coefficient in self-healing mortar with oil core/silica gel shell microcapsules. At early stage (that is, after 3 days), substantial reduction (about 50.2%) was observed for self-healing mortar compared to control specimens; in later stages, the reduction was even higher. Along with carbon microfibers and silica fume, microcapsule material played an important role in reducing permeability. That is, a silica gel shell could participate in cement hydration, and by a physicochemical reaction, helped in the dispersion of carbon microfibers. Smaller fibers in a concrete matrix are often associated with reductions in permeability and therefore joint action of materials and healing agent are effective in reducing the permeability of self-healing mortar.

Other than polymers, carbonate precipitation by bacterial action has proved to be effective in improving durability of concrete. Bacteria encapsulated in diatomaceous earth filled cracks entirely by calcium carbonate precipitation and showed lowest water absorption (Wang et al. 2012a). Wang et al. (2014b) encapsulated bacterial spores with nutrients in melamine-based microcapsules and a water permeability test was done for two different fractions of capsule volumes, namely 3 and 5%. The volume fraction of 3% was deemed optimal from the perspective of effect on mechanical strength and reduced water permeability coefficient. Such a reduction may be attributed to higher carbonate precipitation and pore blocking due to the nutrients added from the rupture of capsules. However, some authors argued that calcium carbonate precipitation could be associated with an increase (e.g., Ngala and Page 1997; St John et al. 1998), or decrease (e.g., Song and Kwon 2007) of porosity, or both, within the same matrix at different locations (Rimmele et al. 2008).

### Repeatability of Self-Healing Action

Regardless of the technique selected, self-healing in concrete must be effective under fatigue loading. In this study, repeatability refers to the performance of self-healing under multiple loading cycles.

Some researchers, including Van Tittelboom et al. (2011b), Thao (2011), and Yang et al. (2011), made efforts to study repeatability of the capsule-based approach using tubular capsules and microcapsules. Van Tittelboom et al. (2011b) tested recovery of strength and stiffness for multiple loading cycles with tubes containing PU as the healing agent. Cracks of about 400 $\mu$m wide were created by load application, although some crack closure took place due to elastic action of mortar and reinforcement bars. Highest average strength recovery of up to about 61% was observed for glass tubes with a 3-mm diameter for the first loading cycle, whereas for second loading, highest recovery of strength was about 23%. Similarly, regained stiffness was higher (about 64%) in first loading but it dropped to maximum 34% for second reloading. Thao (2011) observed similar recovery of stiffness for first and second loading cycles of reinforced beam. Isocyanate prepolymer (epoxy) was encapsulated in glass tubes. Crack width was maintained at around 300 $\mu$m. Maximums of 88 and 85% of normalized stiffness recovery (expressed as fraction of predamage stiffness) were observed for the first and second healing cycle for beams; for columns and slabs, maximum recovered stiffness was 70 and 99%, respectively. Furthermore, under multiple cycles of damage, the repaired cracks did not reopen and new cracks were formed. Yang et al. (2011) studied the fatigue behavior of self-healing mortar with encapsulated MMA in polystyrene microcapsules and reinforced with carbon nanofibers. Fatigue performance was measured in terms of extent of development of micro-strain up to 25,000 uniaxial compressive loading cycles. Self-healing mortar developed smaller microstrains compared to the reference material when subjected to same number of cycles; this, indicates prolonged service life in the self-healing mortar, which can be attributed to increased toughness.

In summary, this review evaluates the effectiveness of different capsule-based self-healing approaches for sustainable infrastructure in terms of the eight effectiveness factors discussed. A comparison of the effectiveness of three encapsulation techniques is provided in Table 5.

## Need for More Sustainability-Related Research

There is generally a lack of work and data on the efficacy of self-healing in an actual application environment. In fact, the aim of these studies should also examine how to enhance the service life and reduce cost, as well as determine the environmental and social benefits brought about by the deployment of self-healing concrete systems. It will also be useful to study how developing self-healing materials to prolong the service life span of civil engineering structures can also contribute to climate change mitigation (Kua and Ashford 2004; Gunawansa et al. 2010; Kua and Koh 2012; Kua and Gunawansa 2010; Gunawansa and Kua 2014; Ng and Mithraratne 2014). Sustainability assessment methods, such as lifecycle assessment, can also be applied to study and improve the

**Table 5.** Comparison of Self-Healing by Different Types of Encapsulation with Respect to Eight Effectiveness Criteria

| Effectiveness criteria | Chemical encapsulation in chemical/ polymer microcapsules | Chemical in glass/ceramic tubes | Encapsulation of bacteria |
|---|---|---|---|
| Robustness during mixing | Can be uniformly mixed and dispersed in the matrix. Capsule wall thickness must be properly designed to prevent premature rupture of capsules | Due to inherent brittleness, some form of protection may be needed, such as mesh, spiral wire, and layer of mortar (Thao 2011) | Can be uniformly mixed and dispersed in the matrix. Capsule wall thickness must be properly determined to prevent premature rupture of capsules |
| Probability of cracks encountering the capsules | Probability of hit may be increased by optimally adjusting size, shape, and dosage of capsules (Lv and Chen 2013) | Glass tubes create weak planes and cracks tend to propagate towards the locations of the tubes<br>Randomly placed tubes are associated with decreased probability of hitting (Van Tittelboom et al. 2015) | Probability of hit may be increased by optimally adjusting size, shape, and dosage of capsules (Lv and Chen 2013) |
| Curing time and condition of healing agent | Curing time and condition depends on the healing agent encapsulated and its viscosity<br>For example, PU with viscosity of 7,200 mPa s may be cured in presence of moisture and take 40–180 min to cure (Van Tittelboom and De Belie 2010) | Curing time and condition depends on the healing agent encapsulated, its viscosity, and number of components | Availability of moisture is a prerequisite for bacterial precipitation (Wang et al. 2014b, a; Jonkers 2011)<br>About 2–3 weeks is needed to seal microcracks (Jonkers 2011; Wang et al. 2012a; Wang et al. 2014a) |
| Effect of empty capsules on concrete strength | Effect on strength depends on capsule material, dosage, and dimension<br>Polyurethane capsules at 2% dosage by weight did not affect strength (Pelletier et al. 2011) | Conventional glass or ceramic tubes create weak plane and reduce strength of concrete<br>Polymeric capsules such as polystyrene are unlikely to adversely affect strength (Hilloulin et al. 2015) | Effect on strength depends on capsule material, dosage and dimension |
| Controllability of release of healing agent | Viscosity of healing agent and capsule shell thickness must be carefully adjusted<br>On average, elongated shape can release higher volume of healing agent compared to spherical shape (Mookhoek 2010) | Due to negative pressure at tube ends, release efficiency of healing agent may be low. Tube dimension is important for efficient release of healing agents (Van Tittelboom et al. 2015) | Capsule shell thickness must be carefully adjusted, so that rupture takes place when crack is intercepted<br>Inadequate data on influence of capsule shape and size when bacteria are encapsulated |
| Stability of healing agent | Can have long shelf life and stability if moisture penetration and trapped bubbles can be avoided | Encapsulation of two-component healing agent increases stability, provided there is proper mixing (Van Tittelboom et al. 2011b) | Using spores rather than live cells ensure stability over longer period of time. So far, results up to 6 months have been studied (Jonkers 2011) |
| Sealing ability and recovery of durability and strength of concrete matrix | Partial recovery in mechanical properties (Pelletier et al. 2011)<br>Substantial reduction in air and gas permeability found (Yang et al. 2011) | Good performance, in terms of crack blocking, recovery of durability and mechanical properties (Van Tittelboom et al. 2015; Thao 2011)<br>Crack sealing effectiveness depends on positioning and protection of tubes (Van Tittelboom et al. 2015; Thao 2011) | Good performance in terms of recovery of durability properties (Wang et al. 2014b, a; Jonkers 2011)<br>Inadequate data on recovery of strength or stiffness |
| Repeatability of self-healing action (under cyclic loading) | Improved fatigue behavior was observed for samples with methyl methacrylate encapsulated in polystyrene microcapsules (size about 4.15 $\mu$m) (Yang et al. 2011)<br>More studies needed to establish consistency | Dependent on protection of glass capsules and ability of healing agent to flow out from tubes<br>Glass capsules were protected by spiral wire coated with mortar with thickness about 3.5 mm. Satisfactory recovery of stiffness in second loading cycle, as shown by Thao (2011) | No published results on performance under multiple loading cycles |

lifecycle environmental impacts of self-healing concrete systems compared to different building materials, including copper slag (Kua 2013a, b), clay bricks (Kua and Kamath 2014), steel slag (Kua 2015), and structural steel of equivalent grades (Kua and Maghimai 2016). Finally, test standards should also be developed to guide applications of self-healing technologies and technique in buildings and other civil engineering structures.

## Conclusion

In the literature, much focus has been placed on autogenous self-healing, but this review showed that self-healing by encapsulation has the potential to deliver higher quality self-healing, in terms of the wider range of crack width that can be healed and faster response to cracking in the matrix.

As discussed, there are different approaches to self-healing by encapsulation, the quality of which can be evaluated in terms of eight effectiveness criteria. These properties also highlight the complexity involved in determining the best combinations for optimum self-healing. At the very least, before selecting a method for a specific intended purpose, factors such as crack width, crack type, nature of the crack formation process (that is, whether it is stabilized or dynamic), and locations of application should be ascertained.

Among other issues, several key challenges for capsule-based self-healing system were identified. Presence of glass capsules and tubes made from ceramic or glass can actually weaken the concrete structure. There is also inadequate data on repeatability of the self-healing process under multiple loading. The situation with encapsulation of bacteria is less clear. Most notably, there is still inadequate data on the influence of capsule shape and size on the effectiveness of self-healing. Furthermore, although survivability of bacteria up to 6 months were shown, for bacteria encapsulation technology to be practical and useful in buildings and civil engineering structures, this survivability has to be prolonged substantially. Finally, there are still inadequate data on recovery of mechanical properties of concrete from bacteria-based self-healing.

One can also conclude from this review that two of the most important factors that influence how efficiently a self-healing technique performs are the crack width and the protection of microcapsules. The volume of healing agent contained in tubes or microcapsules is limited. One of the reasons that self-healing could not be as effective as expected in many of the cases studied is the lack of control over crack propagation. It actually means that a crack developed under loading continues to propagate, thus causing increase of crack width and length even under decreasing loads. Further research must be done to investigate various means of crack width control while employing appropriate self-healing techniques.

Given the development of self-healing materials in the past decade, it may be a practical expectation to soon implement more-widespread application of self-healing materials in buildings. However, before that future can be realized, the highlighted technological and technical issues must first be addressed.

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