CSCE Annual Conference
Growing with youth – Croître avec les jeunes

Laval (Greater Montreal)
June 12 - 15, 2019

SELF-HEALING CONCRETE: A CRITICAL REVIEW

Shannon Guo[1,2], Samir Chidiac[1,3]
[1] McMaster University, Canada
[2] guosx@mcmaster.ca
[3] chidiac@mcmaster.ca

Abstract: This paper presents a comprehensive literature review on the state-of-the-art of autonomous self-healing concrete using microcapsules. Common capsule shell, core materials, and corresponding manufacturing techniques are summarized. The criteria for a successful self-healing system is identified and approaches to enhance the efficiency and effectiveness of self-healing are discussed. The advantages and limitations of autonomous self-healing are identified along with recommendations for future development.

1. INTRODUCTION

Concrete, the world's most used construction material, is susceptible to cracking. Micro-cracks are formed due to shrinkage and thermal expansion at early age, and due to structural loading or environmental actions while in service. Cracks provide entry points for gas, liquid and deleterious chemicals which pose a significant risk to the safety and serviceability of reinforced concrete structures. To mitigate this hazard, self healing materials have emerged as a viable solution. This review paper provides critical analyses of the postulated self-healing materials and procedures for concrete. Topics includes the encapsulation and encapsulated material, manufacturing process, and performance of the microcapsules in concrete.

2. SELF-HEALING

Self-healing mechanisms are categorized as autogenous or autonomous healing. Autogenous healing relies solely on material intrinsic to the cementitious matrix to heal itself. For autonomous healing, foreign elements such as polymers or bacteria are introduced into the concrete to achieve healing. The healing material is typically stored in a vessel embedded in the matrix. Once released from the capsules, the healing agents adhere to the crack faces with the aim to seal openings, recover mechanical properties and prevent further crack propagation.

2.1 Autogenous healing

Autogenous healing is achieved through two mechanisms: continued hydration of unreacted cement particles, and precipitation of calcium carbonate ($CaCO_2$) from the carbonation of calcium hydroxide (Edvardsen 1999). At early ages, autogenous healing is dominated by CSH formation as unreacted cement particles on crack faces are hydrated. At a later stage, owing to insufficient unhydrated cement, healing occurs by precipitation of calcium carbonate. Three conditions must be met for autogenous healing: (1) a continuous supply of water, (2) presence of cementitious minerals to participate in the reactions, and (3) limited crack widths (Snoeck et al. 2014). The healable crack width in the presence of moisture ranges from

20 µm to 138 µm for complete healing and up to 150 µm for partial healing (Yang, Yang, and Li 2011; Snoeck et al. 2014). If the crack is too wide, the cement particles or calcium hydroxide in the surrounding concrete will be consumed before the crack can be completely sealed. Healing effectiveness decreases with concrete age, as reaction products are used up with hydration. Due to the inherent limitations of autogenous healing, a more practical and robust approach to self-healing is needed.

2.2 Autonomous healing

Autonomous self-healing agents are stored in vessels either in the form of a vascular network or dispersed as discrete microcapsules. Targeted release of the encapsulated material in damage zones is achieved through a release trigger such as capsule rupture or change in pH of pore solution. Healing agents react with substances in the internal cementitious environment, chemicals in the host matrix, or with other encapsulated substances, to bind the crack faces together.

A vascular system consists of a network of thin brittle tubes filled with a healing agent and connected to an external reservoir that continuously supplies the healing agent. The healing material is released into the concrete when a crack breaks a section of the tube. While this system has extensive healing potential and good repeatability, a fragile network of brittle tubes is not practical for industrial use due to the additional precautions required to protect it against breakage during casting of concrete (Van Tittelboom and De Belie 2013). A more practical method involves discrete encapsulation of healing agents in spherical or tubular microcapsules. These microcapsules are uniformly dispersed in the concrete matrix and break open when cracks in the concrete propagate through them. Upon rupture, the healing agent is released into the crack through capillary or gravity forces.

3. MICROCAPSULE SHELL MATERIAL

Microcapsules must possess certain properties to remain self healing in concrete. The microcapsule shell material needs to (a) survive the high shear and high temperatures during mixing and curing, (b) afford mechanical release triggers or delay release of the healing agent, (c) be chemically compatible with both the internal and external environment, and (d) preserve the healing potency of the agents by being sufficiently impermeable to prevent leaching of the healing agent and the diffusion of external chemicals into the capsule. Various encapsulating material have been studied.

3.1 Glass and ceramic shell

Glass and ceramic shells are usually limited to tubular capsules due to the manufacturing process. These capsules are relatively inert and easily ruptured, allowing for efficient release of the healing agent in the even of a crack (Thao et al. 2009). Since these capsules have a high risk of cracking during casting, they are manually placed during casting, which is time-intensive and costly. Preventative measures such as metal wire wrapping or precasting in mortar blocks offer limited protection (Tran Diep 2011).

3.2 Polymeric shell

Polymer shells have good versatility, as the fabrication process can be adjusted to obtain desired material properties. Microcapsules are typically made from organic thermosetting resins, such as polyurethane and various amino resins. Polyurethane (PU) is traditionally used for surface coatings to seal concrete from damaging environments, because it adheres well to concrete, has good flexibility and high resistance to ionic permeability. It has been successfully used in the encapsulation of sodium silicate (Pelletier et al. 2010). A hybrid formulation of elastomers forming poly(urea-urethane) (PUU) has also proven to be viable shell material (Tan et al. 2016). Common amino resins used for encapsulation include urea-formaldehyde (UF), melamine-formaldehyde (MF), and phenol-formaldehyde (PF). While UF microcapsules are favoured for its low production cost, it is a poor sealer with lower strength and toughness compared to other amino resins (Sun and Zhang 2002). Copolymers can be fabricated to obtain a cost-effective shell material without negating performance. A recent study on melamine urea-formaldehyde (MUF) microcapsules reported improved encapsulation ability and easier synthesis process compared to UF (Li et al. 2016). Moreover, MUF capsules tend to have thicker robust shells with lower permeability compared to UF (Brown et al. 2003).

3.3 Adaptive materials

Microcapsules shell with mechanical properties that adapts to the surrounding environment may provide an alternative to traditional brittle shell materials. Melamine-based microcapsules are flexible in high humidity but brittle in low humidity (Wang et al. 2014b). As a result, these capsules can withstand the wet mixing process yet can be easily broken when a crack appears in dry concrete. Similar behaviour is observed in gelatin/acacia gum microcapsules, which transition from a soft and 'rubbery' state when hydrated, to a hard and brittle state upon drying (Giannaros, Kanellopoulos, and Al-Tabbaa 2016).

4. HEALING AGENT MATERIAL

Healing agents commonly used in existing literature can be categorized as chemical, mineral, or microbial. Chemical compounds such as epoxy, acrylates, and DCPD rely on polymerization reactions to bind the crack faces together. These healing agents can be further categorized as single- or multi-component. Single-component chemical agents cure on contact with substances in the cementitious matrix or with any moisture or oxygen that enters the crack. They face the risk of premature hardening inside the capsules if external substances diffuse or leak into the shell. For this reason, multi-component systems that store reactive components separately are used for long term stability and prevention of premature activation (Dry and McMillan 1996). Incomplete mixing of components due to uneven release from capsules could reduce the healing efficiency, so healing agents that are insensitive to mix stoichiometry are preferred. Mineral healers, particularly silica-based material, rely on hydration reactions to enhance the intrinsic healing abilities of concrete. Lastly, microbial systems utilize living bacteria to produce healing material.

4.1 Epoxy

Epoxy resin is widely utilized in industrial applications as one of the strongest structural adhesives. Cured epoxy can achieve excellent mechanical and chemical properties, such as high tensile and compressive strength, with good resistance to heat and chemicals. It is often injected in cracks to repair concrete, making it a natural self-healing candidate. Epoxy requires additives to initiate curing, so a curing agent must be mixed in the cement paste. Once the microcapsules crack, epoxy is released into the crack and polymerizes upon contact with curing agent in the cement. For example, Dong et al. (2017) used epoxy encapsulated in UF and premixed a hardener in the concrete. Specimens with 100 µm cracks recovered nearly a third of the initial compressive strength after healing. Epoxy can cure quickly at ambient conditions with amine hardeners but is very sensitive to the mix ratio between epoxy and hardener. When used without hardeners, epoxy is relatively unreactive and requires as much as 7 days to completely cure (Tran Diep 2011).

4.2 Acrylics

Acrylics, such as cyanoacrylate and methacrylate, are a common industrial adhesive. These polymers are valued for their high resistance to chlorides and carbonation. Cyanoacrylates (i.e. ethyl-2-cyanoacrylate, commercially known as super glue) are single component anaerobic adhesives that cure rapidly with minimal moisture. This polymer has low viscosity, which allows it to fill fine microcracks around fracture areas (Joseph et al. 2010). Unlike many other chemical agents, cyanoacrylate is not highly toxic and is commonly used on the human body in dental and medical applications.

Methyl methacrylate (MMA) is traditionally applied as overlays to seal cracks on bridge decks, favoured for its low viscosity, long term storage stability, and extreme temperature tolerance. Unlike cyanoacrylate, methacrylates require a curing agent to initiate polymerization. The curing process is slower than epoxy and cyanoacrylate; as a result, it may be absorbed by the base material. MMA is cured using radical initiators such as benzoyl peroxide (BPO), dimethylparatohidine (DMPT), or triethylborane (TEB). A two-part system consisting of a MMA-DMPT component and MMA-BPO component has been developed with low sensitivity to mix ratio, however skewed proportions tend to delay the initiation of curing (Van Tittelboom et al. 2011).

4.3 Dicyclopentadiene

Dicyclopentadiene (DCPD) is often used as an adhesive or an intermediate to polymer resins. It reacts with a Grubbs catalyst to form a strong crosslinked network within a few minutes at room temperature. DCPD is characterized by low viscosity, low volatility, short curing time, as well as good mechanical properties and resistance to acids and bases after curing. As a multi-component healing agent, DCPD will not cure prematurely within the capsule and has a longer shelf life in sealed capsules. DCPD can be cheaply obtained as a by-product of the petroleum refining process and has relatively low toxicity at low concentrations (Dow Chemical Company 2014). White et al. (2001) first demonstrated the feasibility of DCPD in multi-component self-healing, where DCPD is encapsulated in a UF shell while the Grubbs catalyst is directly mixed into the cement paste. DCPD has since been successfully encapsulated with UF (Brown et al. 2003), MF (Hu, Chen, and Zhang 2009) and PF shells (Lv et al. 2016). Existing studies on the healing effectiveness of DCPD in concrete, however, are severely lacking.

4.4 Silica-containing minerals

While the chemical resins presented thus far have good potential for self-healing of concrete, mineral self-healing compounds have better material compatibility. For example, the benefits of adding silica- or silicate-containing material to concrete has been well documented. Furthermore, silicates are less toxic to human health and the environment, do not require additional chemical curing agents, and are considerably cheaper to obtain compared to polymer resins (Kanellopoulos, Qureshi, and Al-Tabbaa 2015). In encapsulated self-healing applications, sodium silicate has been shown to recover flexural strength, toughness, and improve corrosion resistance (Pelletier et al. 2010). Ethyl silicate (TEOS) can participate in healing through a two-step curing process with water and calcium hydroxide, producing CSH gel (Kanellopoulos, Qureshi, and Al-Tabbaa 2015). Mineral healing agents can be incorporated as a solid or dissolved in a liquid. Aqueous sodium silicate reacts faster and is better at diffusing into cracks than dried crystalline phase, which tends to leave residual healing material in the shell after release (Giannaros, Kanellopoulos, and Al-Tabbaa 2016).

4.5 Bacteria

Crack healing can be achieved using micro-organisms that produce calcium carbonate. Most bacteria cannot survive the harsh alkalinic environment inside concrete and require encapsulation, immobilization in a carrier material, or usage in the form of spores. Bacterial spores remain metabolically dormant until activated by nutrients or water and can survive longer in harsh conditions compared to their active counterparts (Wang et al. 2014b). Wiktor and Jonkers (2011) reported that Bacillus alkalinitrilicus can completely heal cracks of limited width, given sufficient time and supply of water. Bacterial healing showed minimal improvement over autogenous healing in the first 40 days but filled crack widths up to 0.46 mm after 70 days of submersion in water. Bacillus sphaericus spores can achieve up to 80% healing under wet-dry curing conditions (Wang et al. 2014b). Complete healing has been reported for crack widths in the range of 0.15-0.17 mm and up to 0.5 mm (Wang, De Belie, and Verstraete 2012; Wang et al. 2014a). While microbial self-healing is promising due to the environmental sustainability of bacterial agents and long-term healing potential, this method is not practical as water and nutrients must be present to activate and induce high bacterial activity. Furthermore, significant crack healing is only observed after a long healing period, so the healing process may be too slow to prevent crack growth.

5. MANUFACTURING PROCESS

Material selection and manufacturing play a major role in microcapsule performance. Non-polymeric material such as glass and ceramic are used almost exclusively for tubular shells. These capsules are typically produced by filling pre-manufactured hollow glass tubes with healing agent through manual injection, or via capillary action to avoid trapping air bubbles in the tube. Tube ends are sealed with wax or an epoxy that will not react with the encapsulated material or cementitious material surrounding the capsule (Joseph et al. 2010; Thao et al. 2009). Polymer microcapsules have the advantage of mechanical properties that can be optimized by altering the fabrication procedure. Many existing microcapsule polymerization techniques are well established and frequently used in the food, cosmetic, and pharmaceutical industry.

Polymer microcapsules used in concrete applications are commonly synthesized via chemical microencapsulation techniques such interfacial or *in-situ* polymerization. Spherical polymer capsules produced using interfacial polymerization may be the most suitable for commercialization. Interfacial polymerization offers fast encapsulation, high encapsulation efficiency, milder reaction conditions, and is less sensitive to reactant stoichiometry and purity. In-situ polymerization also offers good control of capsule properties at a low cost and simple synthesis process, despite the reaction requiring a longer time compared to interfacial polymerization (Zhu, Rong, and Zhang 2015).

6. EVALUATING A SELF-HEALING SYSTEM

As a foreign element in the host matrix, microcapsules affect both the wet and hardened concrete properties. Accordingly, the ideal self-healing composite must optimize self-healing efficiency and effectiveness without compromising the original material properties. The efficiency of an encapsulated healing system depends on capsule morphology, shell composition, and the encapsulated content, whereas effectiveness is evaluated through the recovery of strength and durability after healing.

6.1 Microcapsule shell robustness & survivability

Microcapsule robustness can be defined as the ability to withstand extreme stresses and external conditions without losing the desired functionality. To ensure the healing agent can be readily released into the damage zone when a crack develops, microcapsules must be mechanically, thermally and chemically robust. Most importantly, mechanical robustness is necessary to ensure healing agents are not prematurely released. This is because self-healing microcapsules are typically dispersed in the mix water during concrete preparation. As capsule shells are made from brittle material, microcapsules are prone to damage or rupture from collisions and high shear stresses imposed by mechanical mixing.

Mechanical robustness is affected by physical characteristics of the microcapsule shell. In particular, maximum burst load for a microcapsule depends on both the shell thickness and capsule diameter. Microcapsules with a low thickness-to-diameter ratio offer greater storage capacity for a given volume, but risk premature rupture and diffusion of material through the shell. A higher ratio of shell material enhances the mechanical properties for survival during mixing, but may impede mechanical release triggers (Li et al. 2016). Shell morphology also has a major effect on capsule robustness, particularly when a brittle material are selected for the shell (Lv, Li, and Chen 2017). Long narrow tubes and microvascular systems require careful preparation and protection during setting to avoid damage. Common protection practices include binding tubular capsules to reinforcement bars, the placing of a layer of mesh reinforcement above the capsules, and encasement in cement mortar or wire wrappings (Thao et al. 2009). Discrete spherical microcapsules are significantly easier to incorporate into the concrete but provide a limited volume of healing material. Elongated spherocylindrical capsules may be able to combine the large storage capacity of tubular capsules with the practicality of spherical capsules (Mookhoek 2010).

6.2 Release efficiency

Inefficient release of the healing agent is detrimental as it could lead to delayed healing and further crack growth. Upon crack development, microcapsules are expected to promptly deploy the healing material in response to a trigger. The most common release mechanism is the mechanical rupturing of microcapsules from stresses caused by cracking the concrete. The main disadvantage of this type of release trigger is the difficulty in optimizing microcapsule properties for both shell survival and response to mechanical stresses.

6.2.1 Rupture behaviour

Mechanical rupture depends on two conditions: (1) probability of a crack passing through a microcapsule, and (2) sensitivity of the microcapsule to crack stresses. Numerical models suggest that the probability can be calculated based on capsule morphology and quantity in the bulk medium (Zemskov, Jonkers, and Vermolen 2011). High aspect ratio morphologies such as tubular shapes have a higher probability of intersecting the crack plane. Furthermore, for a given capsule dosage and volume, elongated capsules can deliver more healing agent to the crack surface (Lv, Li, and Chen 2017). Unfortunately, the practicality of cylindrical capsules is greatly restricted by its shape, as high release efficiency requires capsules to be placed perpendicular to the fracture plane.

Mechanical properties such as microcapsule strength and stiffness also affect crack propagation behaviour in the vicinity of the capsule (Lv et al. 2017). An inclusion with higher elastic modulus than the surrounding matrix creates a stress field that deflects cracks away from the inclusion, while inclusions with low elastic modulus will attract cracks (White et al. 2001). Once a crack successfully hits a microcapsule, tensile stresses concentrated at the crack tip must break open the capsule to facilitate release. Strong interfacial bonding between capsule shell and cementitious matrix is necessary to ensure the crack does not propagate along the interface, which would debond the capsule and prevent rupture. Numerical models suggest that debonding is governed by the bond strength, strength and stiffness of capsules relative to the matrix, as well as the thickness-to-diameter ratio of microcapsules (Gilabert, Garoz, and Van Paepegem 2015). While it is realistically impossible to achieve perfect bonding between capsule shell and cementitious matrix, a rough microcapsule surface can improve bonding by increasing mechanical interlocking and contact area for bonding to take place. This can be optimized in certain polymeric shells by increasing surface depositions (Li et al. 2016) or using high molecular weight shell prepolymers (Dunker et al. 1986). Alternatively, it is possible that strong bonds can be induced with the use of pozzolanic shell material, however this has not been extensively investigated. (Yang et al. 2011) suggested that silica-containing shells can react with calcium hydroxide in the concrete to form a tight shell-cement interface.

6.3 EFFECT ON BULK PROPERTIES

As microcapsules are not a load-bearing aspect of the matrix, the volume taken up by microcapsules are equivalent to capsule-shaped voids that can adversely affect the concrete strength. Compressive strength drops significantly when inclusions exceed 3 wt%. Twenty eight day compressive strength of specimens containing 5-180 µm silica capsules at 5 wt% and 10 wt% experienced around 36% reduction in strength (Perez et al. 2015). Dong et al. (2017) reported 25% decrease in compressive strength with 8 wt% dosage of 180 µm UF capsules. For the same volume fraction, inserting larger capsules at a lower quantity of capsules may reduce the loss in strength (Kanellopoulos, Giannaros, and Al-Tabbaa 2016). Furthermore, it may be beneficial to only add microcapsules in potential cracking zones to maximize healing potential without sacrificing compressive strength. Elastic modulus and tensile strength are not significantly impacted by the addition of microcapsules (Dong et al. 2017; Wang et al. 2014b).

6.4 HEALING EFFECTIVENESS

Efficient self-healing entails recovery of concrete’s mechanical and transport properties close to that of the undamaged specimen. Durability of damaged concrete is typically recovered through physical sealing of the crack, which prevents ingress of liquids, solids, or gases. Meanwhile, mechanical recovery depends on the strength of the healing material that is binding the crack surfaces.

6.4.1 Recovery of durability

By reducing the permeability of the damage zone through sealing of cracks or densification of the damaged cementitious matrix, the durability of cracked concrete can be improved. Dong et al. (2016) observed a reduction in capillary porosity, continuous pore diameter, pore connectivity, and reduced chloride penetration in healed mortar specimens containing epoxy capsules. For the same amount of healing material, larger microcapsules are more effective than small microcapsules (Rule, Sottos, and White 2007). For example, the use of 230 µm capsules improved impermeability by approximated 22%, while the same weight fraction of 132 µm capsules decreased impermeability by only 14% (Dong et al. 2017). Sodium silicate exhibits greater sealing effectiveness than certain polymers. In specimens containing 6 wt% of 290 µm sodium silicate microcapsules, complete sealing is observed in 110-170 µm wide cracks and 80% sealing in 180-250 µm cracks (Kanellopoulos, Giannaros, and Al-Tabbaa 2016). In comparison, only 33% and 26% closure, respectively, of similar crack openings are observed when 6 wt% of 230 µm epoxy microcapsules are used (Dong et al. 2016). The effect of healing may be reduced if there are aggregates surrounding the microcapsules, since healing agents such as sodium silicate have limited reactivity with non-cementitious aggregates (Kanellopoulos, Giannaros, and Al-Tabbaa 2016).

6.4.2 Recovery of mechanical properties

Several studies have confirmed that the binding ability of both polymer and mineral healing agents, however the healing capacity is insufficient to restore more detrimental damage caused by large structural cracks. Specimens with 100-300 µm cracks experienced only 6.5-13% recovery in compressive strength after healing (Dong et al. 2016). Around 20% recovery of flexural strength can be recovered with DCPD capsules (Gilford et al. 2014) and 20-26% recovery with sodium silicate capsules (Pelletier et al. 2010).

7. LIMITATIONS OF CURRENT STATE OF RESEARCH

At present, the application of self-healing technology is not consistent in part due to unsystematic reporting of information in addition to the lack of standardized testing and selection criteria. To facilitate the optimization of self-healing systems, the aim and scope of healing must be defined as a priori. This entails identifying (1) the targeted concrete property to be recovered in healing, (2) the targeted crack type to be healed, (3) test methods that realistically simulate the cracks initiation and propagation, and (4) relevant test methods to quantify the recovery of the targeted concrete property.

‘Self-healing’ of concrete is made up of two concurrent concepts: self-sealing and self-healing of cracks. Sealing requires plugging of the openings, whereas healing refers to recovery of mechanical properties. It is important to differentiate between these two concepts, as the requirements for a durability-based design and strength-based design are different. Furthermore, depending on the purpose of the concrete structure or exposure conditions, recovery of one property may be more advantageous than others. For example, concrete structures exposed to chlorides will benefit more from crack sealing than crack healing, while interior structures will for most cases not benefit from crack sealing. Accordingly, the selection and optimization of microcapsules depend on the prescribed healing.

Crack widths range from a few microns to several centimeters. It is impractical to design microcapsules that heal cracks with wide range of length scales; therefore, the scope of healing should be focused to target a specific crack width. For example, early age concrete is susceptible to cracking due to autogenous, drying, and thermal shrinkage. Compared to drying shrinkage and thermal volume changes, deformations due to autogenous shrinkage are significantly smaller (Jensen and Hansen 2001). Drying-induced microcracks are usually less than 0.1 mm in width and with limited penetration depth up to 18 mm (Wu, Wong, and Buenfeld 2015). Temperature gradients caused by poor dissipation of heat from hydration can induce microcracks in the range of 0.01-0.1 mm and penetration depth less than 50 mm (Delatte 2009). Concrete is also susceptible to structural cracks from loading throughout its service life. These types of cracks have highly variable sizes, depending on the service load. For reinforced flexural members, CSA A23.3-04 limits the allowable crack width to about 0.4 mm for interior exposure and 0.33 mm for exterior exposure. In comparison, microcapsules appear to be most suitable for healing of microcracks no wider than 0.2 mm. In order to achieve good healing, it is advisable for the self-healing system to target the most critical crack.

To accurately assess the healing potential, it is necessary to simulate cracks that are representative of realistic crack mechanisms. Crack characteristics will vary depending on the testing method, yet most studies test self-healing without a basis for the test method chosen to induce cracking. Cracks are typically created using common techniques such as the compression test, indirect tensile test, and flexural bending. Three-point and four-point bending generate V-shaped cracks that are wide at the crack opening but taper to a tip at the end of the crack. In contrast, split tensile tests produces cracks with more uniform width along its length. In practice, three- or four-point bending cracks are representative of flexural cracks or shrinkage cracks, whereas tensile tests may reproduce internal cracks caused by restrained deformations (Ferrara et al. 2018).

Due to the lack of standardization, many studies utilize different methods and diverse evaluation criteria to assess self-healing. Recovery of mechanical strength is typically assessed by comparing mechanical properties between pre-healing and post-healing specimens, or between healed specimens and non-healed specimens under the same curing conditions. The recovery of durability is measured in terms of liquid permeability, gas permeability, sorptivity, or chloride diffusion. While these measures provide a comparative assessment, they do not provide a comprehensive and consistent metric for quantifying the efficiency and effectiveness of healing systems.

8. CONCLUDING REMARKS

This review has revealed that the use of microcapsules for self-healing in concrete has the potential to maintain the structural integrity and longevity of concrete structures. The challenges are three folds: Self-healing system negatively impacts the rheological, mechanical and physical properties of concrete; Self-healing system is expected to heal a wide range of length scales; Self-healing system raises production costs. Nonetheless, self-healing concrete can afford the concrete industry to become sustainable; environmentally, societally and economically. Self-healing concrete will mitigate premature failures; increase durability and service life; reduce traffic and living disruptions; and reduce utilization of energy, non-renewable materials and demand for landfill.

Issues preventing the wide-spread implementation of self-healing concrete have been identified. Accordingly, there is a need to develop standardized approach for selecting, designing and testing self-healing systems in concrete. Moreover, a comprehensive experimental and analytical studies are recommended for investigating the physical, mechanical, thermal and chemical requirements for the microcapsules to be compatible with the wet and hardened concrete.

9. REFERENCES

Brown, E.N., Kessler, M.R., Sottos N.R., and White S.R. 2003. In Situ Poly(Urea-Formaldehyde) Microencapsulation of Dicyclopentadiene. *Journal of Microencapsulation,* **20**(6): 719–30.

Delatte, N. 2009. *Failure, Distress and Repair of Concrete Structures*.

Dong, B., Guohao F., Weijian D., Yuqing L., Jianchao Z., Ningxu H., and Feng X. 2016. Self-Healing Features in Cementitious Material with Urea–formaldehyde/Epoxy Microcapsules*. Construction and Building Materials* **106**(March): 608–17.

Dong, B., Guohao F., Yanshuai W., Yuqing L., Shuxian H., Jianchao Z., Shangmin L., and Feng X. 2017. Performance Recovery Concerning the Permeability of Concrete by Means of a Microcapsule Based Self-Healing System. *Cement and Concrete Composites* **78**(April): 84–96.

Dow Chemical Company. 2014. Product Safety Assessment: Dicyclopentadiene (DCPD).

Dry, C. and McMillan W. 1996. Three-Part Methylmethacrylate Adhesive System as an Internal Delivery System for Smart Responsive Concrete. *Smart Materials and Structures* **5**(3): 297–300.

Dunker, A.K., John W.E., Rammon R., Farmer B., and Johns S.J. 1986. Slightly Bizarre Protein Chemistry: Urea-Formaldehyde Resin from a Biochemical Perspective. *The Journal of Adhesion* **19**(2): 153–76.

Edvardsen, C. 1999. Water Permeability and Autogenous Healing of Cracks in Concrete. *ACI Materials Journal* **96**(4): 448–54.

Ferrara, L., Van Mullem, T., Alonso, M.C., Antonaci, P., Borg, R.P., Cuenca, E., Jefferson, A., Ng, P.-L., Peled, A., Roig-Flores, M., Sanchez, M., Schroefl, C., Serna, P., Snoeck, D., Tulliani, J.M., and De Belie, N. 2018. Experimental characterization of the self-healing capacity of cement-based materials and its effects on the material performance: A state of the art report by COST Action SARCOS WG2. *Construction and Building Materials* **167**: 115–142.

Giannaros, P., Kanellopoulos, A., and Al-Tabbaa A. 2016. Sealing of Cracks in Cement Using Microencapsulated Sodium Silicate. *Smart Materials and Structures* **25**(8): 084005.

Gilabert, F.A., Garoz, D., and Van Paepegem W. 2015. Stress Concentrations and Bonding Strength in Encapsulation-Based Self-Healing Materials. *Materials and Design* **67**: 28–41.

Gilford, James, Hassan, M.M., Rupnow T., Barbato M., Okeil A., and Asadi, S. 2014. Dicyclopentadiene and Sodium Silicate Microencapsulation for Self-Healing of Concrete. *Journal of Materials in Civil Engineering* **26**(5): 886–96.

Hu, J., Chen, H.Q., and Zhang, Z. 2009. Mechanical Properties of Melamine Formaldehyde Microcapsules for Self-Healing Materials. *Materials Chemistry and Physics* **118**(1): 63–70.

Jensen, O.M., & Hansen, P.F. 2001. Autogenous deformation and RH-change in perspective. *Cement and Concrete Research* **31**(12): 1859–1865.

Joseph, C., Jefferson, A.D., Isaacs, B., Lark, R., and Gardner, D. 2010. Experimental Investigation of Adhesive-Based Self-Healing of Cementitious Materials. *Magazine of Concrete Research* **62**(11): 831–43.

Kanellopoulos, A., Giannaros, P., and Al-Tabbaa, A. 2016. The Effect of Varying Volume Fraction of Microcapsules on Fresh, Mechanical and Self-Healing Properties of Mortars. *Construction and Building Materials* **122**: 577–93.

Kanellopoulos, A., Qureshi, T.S., and Al-Tabbaa, A. 2015. Glass Encapsulated Minerals for Self-Healing in Cement Based Composites. *Construction and Building Materials* **98**(November): 780–91.

Li, W., Zhu, X., Zhao. N., and Jiang, Z. 2016. Preparation and Properties of Melamine Urea-Formaldehyde Microcapsules for Self-Healing of Cementitious Materials. *Materials* **9**(3): 152.

Lv, L., Yang, Z., Chen, G., Zhu, G., Han, N., Schlangen, E., and Xing F. 2016. Synthesis and Characterization of a New Polymeric Microcapsule and Feasibility Investigation in Self-Healing Cementitious Materials. *Construction and Building Materials* **105**(February): 487–95.

Lv, L., Zhang, H., Schlangen, E., Yang, Z., and Xing, F. 2017. Experimental and Numerical Study of Crack Behaviour for Capsule-Based Self-Healing Cementitious Materials. *Construction and Building Materials* **156**(December): 219–29.

Lv, Z., Li, S., and Chen, H. 2017. Analytical Model for Effects of Capsule Shape on the Healing Efficiency in Self-Healing Materials. *PLOS ONE* **12**(11): e0187299.

Mookhoek, S.D. 2010. *Novel Routes to Liquid-Based Self-Healing Polymer Systems*.

Pelletier, M.M., Brown, R., Shukla, A., and Bose, A. 2010. Self-Healing Concrete with a Microencapsulated Healing Agent., Master's thesis, University of Rhode Island.

Perez, G., Gaitero, J.J., Erkizia, E., Jimenez, I., and Guerrero, A. 2015. Characterisation of Cement Pastes with Innovative Self-Healing System Based in Epoxy-Amine Adhesive. *Cement and Concrete Composites* **60**: 55–64.

Rule, J.D., Sottos, N.R., and White, S.R. 2007. Effect of Microcapsule Size on the Performance of Self-Healing Polymers. *Polymer* **48**(12): 3520–29.

Snoeck, D., Van Tittelboom, K., Steuperaert, S. Dubruel, P. and De Belie, N. 2014. Self-Healing Cementitious Materials by the Combination of Microfibres and Superabsorbent Polymers. *Journal of Intelligent Material Systems and Structures* **25**(1): 13–24.

Sun, G. and Zhang, Z. 2002. Mechanical Strength of Microcapsules Made of Different Wall Materials. *International Journal of Pharmaceutics* **242** (1–2): 307–11.

Tan, N. P. B., Keung, L. H., Choi, W. H. Lam, W. C. and Leung, H. N. 2016. Silica-Based Self-Healing Microcapsules for Self-Repair in Concrete. *Journal of Applied Polymer Science* **133** (12).

Thao, T.D. P. Johnson, T. J. S. Tong, Q. S. and Dai, P. S. 2009. Implementation of Self-Healing in Concrete – Proof of Concept. *The IES Journal Part A: Civil & Structural Engineering* **2** (2): 116–25.

Van Tittelboom, K., Adesanya, K., Dubruel, P., Van Puyvelde, P. and De Belie, N. 2011. Methyl Methacrylate as a Healing Agent for Self-Healing Cementitious Materials*. Smart Materials and Structures* 20 (12): 125016.

Van Tittelboom, K. and De Belie, N. 2013. Self-Healing in Cementitious Materials—A Review. *Materials* **6** (6): 2182–2217.

Wang, J., De Belie, N. and Verstraete W. 2012. Diatomaceous Earth as a Protective Vehicle for Bacteria Applied for Self-Healing Concrete. *Journal of Industrial Microbiology and Biotechnology* **39**(4): 567–77.

Wang, J., Snoeck, D., Van Vlierberghe, S., Verstraete, W. and De Belie, N. 2014a. Application of Hydrogel Encapsulated Carbonate Precipitating Bacteria for Approaching a Realistic Self-Healing in Concrete. *Construction and Building Materials* **68**: 110–19.

Wang, J., Soens, H., Verstraete, W. and De Belie. N. 2014b. Self-Healing Concrete by Use of Microencapsulated Bacterial Spores. *Cement and Concrete Research* **56**: 139–52.

White, S. R., Sottos, N.R., Geubelle, P. H., Moore, J. S., Kessler, M. R., Sriram, S. R., Brown, E. N. and Viswanathan. S. 2001. Autonomic Healing of Polymer Composites. *Nature* **409** (6822): 794–97.

Wiktor, V. and Jonkers. H.M. 2011. Quantification of Crack-Healing in Novel Bacteria-Based Self-Healing Concrete. *Cement and Concrete Composites* **33** (7): 763–70.

Wu, Z., Wong, H.S. and Buenfeld. N.R. 2015. Influence of Drying-Induced Microcracking and Related Size Effects on Mass Transport Properties of Concrete. *Cement and Concrete Research* **68**: 35–48.

Yang, Y., Yang, E.H. and Li, V.C. 2011. Autogenous Healing of Engineered Cementitious Composites at Early Age. *Cement and Concrete Research* **41** (2): 176–83.

Yang, Z., Hollar, J., He, X. and Shi, X. 2011. A Self-Healing Cementitious Composite Using Oil Core/Silica Gel Shell Microcapsules. *Cement and Concrete Composites* **33** (4): 506–12.

Zemskov, S.V., Jonkers, H. M. and Vermolen, F. J. 2011. Two Analytical Models for the Probability Characteristics of a Crack Hitting Encapsulated Particles: Application to Self-Healing Materials. *Computational Materials Science* **50** (12): 3323–33.

Zhu, D.Y., Rong, M.Z. and Zhang, M. Q. 2015. Self-Healing Polymeric Materials Based on Microencapsulated Healing Agents: From Design to Preparation. *Progress in Polymer Science* **49**: 175–220.