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TECHNICAL PAPER

A review study on encapsulation-based self-healing for cementitious materials

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Encapsulation-based self-healing technology is an effective method for healing the crack-deteriorated cementitious material. Encapsulation-based self-healing initiates by crack occurrence and progresses by chemical reaction of released self-healing agents in the cracks, which are contained in capsules. In this paper, a review has been conducted on various healing agents, encapsulation techniques, as well as experimental approaches, basing on existing substantial studies. Recently, there is no consistent agreement on the effective criteria for evaluating encapsulation-based self-healing and mature solution for increasing the survival ratio of capsules during mixing. However, the polyurethane-based healing agents filled in glass or ceramic tubes are popularly applied for self-healing cementitious materials. Besides, the polymer capsules present promising attractions for engineering application. Mechanical strength and durability are the most widely used self-healing efficiency assessment indexes. On the other hand, nondestructive technique and numerical modeling have also extensively adopted to visualize and evaluate the self-healing behavior of cementitious materials. However, there are still some challenges, which require further investigations, such as behavior of crack propagation, kinetics of healing agent in discrete crack surfaces, effect of inserted capsules on the mechanical properties of self-healed cementitious materials.

KEYWORDS
cementitious materials, crack, efficiency, encapsulation agent, self-healing

1 | INTRODUCTION

Concrete is the most essential and popular material for built infrastructures, due to high serviceability, compress strength, and availability of raw materials.[1] However, the mechanical strength and durability of concrete structures can be seriously deteriorated by micro or macro-cracks.[2] Typically, penetrations of water and chloride through the cracks drastically affect the durability of concrete structures and cause the corrosion of steel bars, which consequently lead to failure issues.[3] Therefore, it is vital to heal or repair these cracks to maintain the serviceability of concrete structures.[4] Cracks can be repaired manually in rare situations, because most of these repairing operations are restricted by accessibility, location, cost, and environment problems.[5] Hence, self-healing of cracks has become a necessity for concrete structures, and attracts rising attention of researchers. On the other hand, the autogenous repairing phenomenon has been observed in natural environment for many years.[6,7] It is accomplished by formation of calcium carbonate and continuous hydration of anhydrate cementitious materials,[6,8] when water and carbon dioxide are available.[9] However, without further modification or improvement, the efficiency of those healing activities is relatively low. Therefore, various strategies have been developed to improve self-healing behavior,[10] typically though inserting kinds of additions into the cementitious materials, such as crystalline admixture,[11–14] polymers,[15,16] hollow fiber,[17] mineral admixtures,[18,19] encapsulations,[20–25] nanoparticles,[26] and microorganisms.[27] In addition, engineered cementitious composite (ECC) with unique micro-crack behavior and tight

Discussion on this paper must be submitted within two months of the print publication. The discussion will then be published in print, along with the authors' closure, if any, approximately nine months after the print publication.

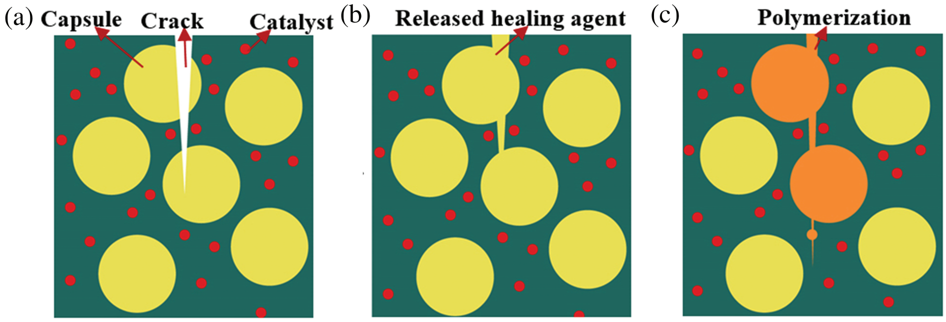

FIGURE 1 Self-healing schematic concept for cementitious materials[10]: (a) crack forming; (b) healing agent releasing; (c) polymerization with catalysts

crack width control properties also shows great potential in crack self-healing.[28,29] Among all these available self-healing concrete technologies, encapsulation-based self-healing concrete is considered as the most promising one, when simply taking the maximum sealed crack width and depth as self-healing governing parameters, according to the comparison made by Muhammad et al.[2] Moreover, the healing process of encapsulation strategy is more flexible, with no need for additional water, less curing time, and repeatable seal-healing actions comparing with other traditional approaches.[24,30,31]

Encapsulation-based self-healing cementitious material originated from the research conducted by White et al.,[15] a kind of crack-self-healing material that incorporated an embedded microencapsulation was proposed. This sort of microencapsulation releases some healing agent that can polymerize with embedded catalyst to seal and heal the cracks when it is broken by crack intrusion, as illustrated in Figure 1. After that, encapsulation-based self-healing technology has been extensively introduced for cementitious materials, and is recognized as the most promising way to achieve self-healing of concrete infrastructures. Common configuration of encapsulation-based self-healing concrete specimens is shown in Figure 2. Some microcapsules are embedded in the cement matrix. These capsules are arranged perpendicular to the precrack in experiments, and usually closely coupled with other capsules containing accelerator or water to investigate the effects of additives on the self-healing process.[32] The shells of these capsules are usually fabricated from brittle materials, and the shape of concrete specimens can be prism, cylinder, or dog-bone, depending on the adopted experiment methods.

The healing principle of encapsulation-based self-healing concrete heavily relies on the immediate rupture of prior inserted capsules, the flow of internal healing agents into crack location and the following adequate curing reactions.[33] Therefore, to guarantee high efficient self-healing, these three sub-processes must be promptly activated.[34] There are various factors which influence the curing process of encapsulation-based self-healing concrete, including the composition of healing agents,[35–39] the capsule materials,[21,40–43] accelerators,[21,32] ambient conditions,[44,45] as well as crack patterns.[21,30,46,47] Besides, experimental methods for self-healing efficiency assessment also affect results.[7,21,30,32] The aim of this review is to discuss different parameters which are relevant to the efficiency of encapsulation-based self-healing cementitious material, and propose valuable suggestions for the future investigation on this topic.

2 | ENCAPSULATION-BASED SELF-HEALING AGENTS

Inspired by the self-repair polymeric materials, most of healing agents in encapsulation-based self-healing concrete are

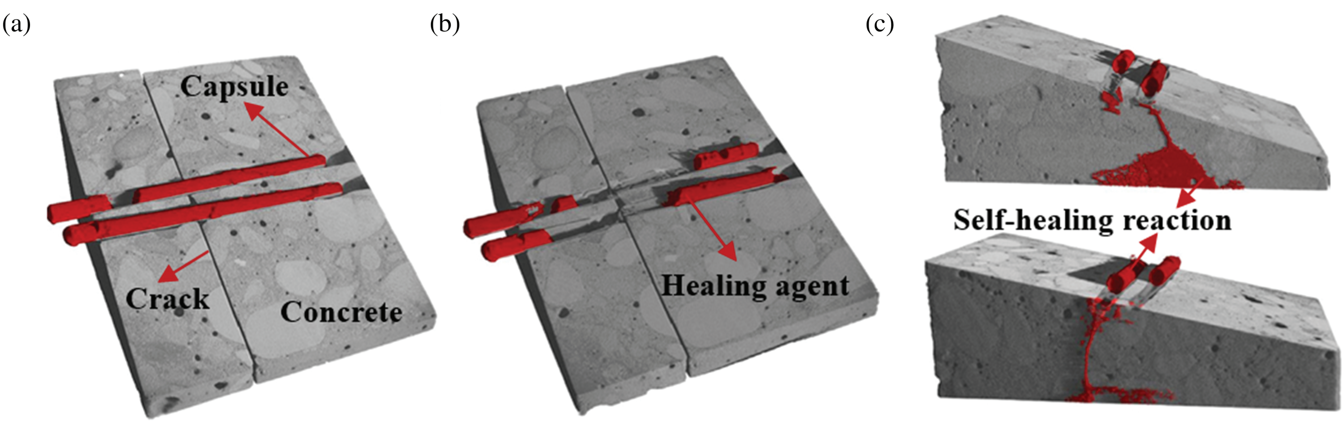

FIGURE 2 Configuration of encapsulation-based self-healing concrete[61]: (a) crack breaking capsule; (b) healing agent filling; (c) healing agent curing

TABLE 1 Different types of self-healing agents for cementitious materials

Self-healing agents	Crack width	Curing time	Mechanical recovery	Self-healing mechanism	Disadvantages	References
Single component						
Cyanoacrylate	<100 μm	Seconds	—	Rapid anionic polymerization after tracing hydroxide ions	Requirement for water and oxygen; healed cracks were limited to 100 μm	40,49,51
Epoxy resin	300 μm	100 min	30%	Harden when heated or pressurized	High viscosity; long curing time	31,60
Polyurethane	250 μm	—	35–80%	Polymerization in moist surrounding	Stress concentration at the interface; detachment from the crack face	30,58,79,84
Sodium silicate	40 μm	—	20–26%	Reaction with calcium hydroxide to produce C-S-H gels	Low mechanical recovery ability	53
Multicomponents						
Methyl methacrylate	—	24 hr	—	Thermally stimulated molecular inter-diffusion	Premature absorption by matrix	57,58,62,71,108
Polyurethane	225–300 μm	—	35–80%	Enhanced polymerization in moist surrounding	Premature reaction	21,32,47,63,66,69
Epoxy resin	225–300 μm	24 hr	35–80%	Accelerated harden with lower viscosity	Insufficient mixing with hardener	37,44,47,60,61

polymers,[21,30,32,48,49] because polymers are endowed with potential capacities for cracks healing, such as immediate polymerization,[15] low viscosity,[32] and stress transfer capability.[48] Reaction mechanism and behavior of healing-agents are directly related to the healing-efficiency of encapsulation-based self-healing technologies.[50] Therefore, developing perfect healing agent becomes the priority in the studies on encapsulation-based self-healing concrete. The healing agents employed by encapsulation-based self-healing concrete are summarized in Table 1, which are mainly divided into two categories: (a) single component healing agents; and (b) multicomponents healing agents. As for multicomponents healing agents, premature chemical reactions between the accelerators and healing agents before flowing into cracks may be the main restriction for the engineering applications.[21,51]

2.1 | Cyanoacrylate healing agent

The cyanoacrylate contained in hollow glass fibers is the early used self-healing agent as superglue to seal cracks for cementitious material. Li et al.[40] reported that the self-healing effectiveness of cyanoacrylate was measured by the elastic modulus regaining of ECC reinforced beams in repeat loading, but the thickness of healed cracks were limited to 50 μm. Although when the moisture and oxygen were available, the low viscosity of healing agent facilitated the rapid curing process (in seconds), the width of crack healing ability was confined to less than 100 μm, due to the required capillary suction forces.[51] On the other hand, short setting time may induce insufficient dispersion of the healing agent within the cracks, leaving unreacted cyanoacrylate in capsules. However, Lark et al.[49] suggested that the left cyanoacrylate could remain liquid state for more than a week, and might attribute to a tertiary healing effect, which was also confirmed by Gardner et al.[51] Compared with other healing agents, the great bonding strength between cured cyanoacrylate and the crack surfaces can prevent new crack formations during reloading, and the curing process can be accelerated by the alkaline environmental condition.[40,49,51]

2.2 | Sodium silicate solution healing agent

Sodium silicate solution reacts with the calcium hydroxide in cementitious materials and produces calcium silicate hydrates (C-S-H) gels for cracks healing. Gilford III et al.[52] reported that an improvement of 11% in elasticity modulus wad achieved for the concrete after being healed by sodium silicate filled in the microcapsules. Mostavi et al.[53] developed a double-walled microcapsule filled with sodium silicate, and put additional focus on the performance of the capsules. As the self-healing efficiency is affected by concentration of sodium silicate solution, the exhaustion of healing agents and low mechanical strength may restrict the engineering application of sodium silicate solution.[54,55]

2.3 | Methyl methacrylate-based healing agent

Methyl methacrylate (MMA) was selected as healing agent by few researchers. Dry[56] realized the successful release of MMA from fibers into white cement during heating, and obtained desirable results in the water permeability test. Yang et al.[57] microencapsulated MMA in silica gel shell to develop a new type of self-healing materials, and found improvement in the gas permeability test. However, when Tittelboom et al.[58] filled the MMA in borosilicate capillary glass tubes, and obtained no improvement compared to the untreated cracks in water ingress test. The reason may be the premature curing of the healing agent in the capsules.

2.4 | Epoxy resin-based healing agent

Epoxy resin is another early used healing agent, which can harden by heating or pressurization.[59] Thao et al.[31] incorporated epoxy resin in a steel-mesh-reinforced mortar specimen by embedding glass tubes, and obtained an increase in strength approximately by 30% compared to the initial strength after repeated autonomic healing. In a complex heating self-healing system for concrete proposed by Nishiwaki et al.,[60] although high self-healing outcome was yeilded by accelerated hardening reaction of epoxy resin, the curing process still took nearly 100 min, due to high viscosity of this kind of healing agent. Therefore, recent investigations related to epoxy resin healing agent provides an additional focus on reducing curing time by introducing accelerators, which is termed as multiple-component epoxy resin healing agent. Li et al.[61] achieved higher healing efficiency in polymer, using polyether amine as hardener of healing agents. On the other hand, some researchers optimized the healing process by diluting this kind of epoxy resin. Tittelboom et al.[62] mixed epoxy with MMA to develop one sort of healing agent with lower viscosity, and found obvious enhancement in the water permeability. Besides, they also combined polyurethane (PU) and epoxy resin from two separate tubes as two-component healing agents to accelerate the polymerizing reaction. Thao et al.[42] proved that the epoxy polymer with low viscosity between 250 and 500 mPa s could flow smoothly into cracks and provide efficient healing, but the specific viscosity still requires further investigations.

2.5 | PU-based healing agent

PU-based healing agent accomplishes crack-healing process by foaming and expanding chemical reaction. Therefore, a little healing agent is enough for large crack healing, since the expansion can seal larger cracks.[21] Tittelboom et al.[30] proved that the moisture in the cementitious materials was sufficient for the polymerization reaction, which made PU a versatile healing agent. They also found that more than 50% increase in the original strength and stiffness can be regained. The water permeability could be greatly reduced after self-healing by PU,[21] which was also confirmed by the neutron radiography visualization results.[58] Maes et al.[63] found that PU is able to reduce chlorides penetration along the crack path by 67 and 33% for initial cracks with widths of 100 and 300 μm, respectively. The self-healing efficiency and advantages of PU healing agent have been verified by many studies.[46,63] Therefore, current investigations on PU-based healing agent primarily aim at determining the specific parameters to obtain the excellent healing performances, such as accelerator, suitable viscosity, stiffness, elastic modules, strain capacity, bonding strength, and interface properties.

Feiteira et al.[32] analyzed the strain capacity of cured PU-based polymers, considering elongation deformation of fatigue cracks under cyclic load.[64,65] It was found that the PU-based healing agent with super low viscosity (200 mPa s) can meet the strain capacity requirement ranging from 50 to 100%, and reduce the failures caused by breakage of foam structures, which was also confirmed by Feiteira et al.[66] Moreover, they suggested that flexible polymers with elastic modules much lower than 10 MPa could withstand crack propagation, prevent initiation of new cracks, and decrease interface stress, thus reduce the detachment of the bonding. However, Dry et al.[48] found that the relative stiffness of an adhesive healing agent might affect the repairing ability. They suggested that low stiffness might cause poor regained stiffness values, but stiff adhesive can easily transfer stress across cracks, allowing the crack to develop continuously. On the other hand, Gilabert et al.[67] evaluated the strength contribution of cured PU-based healing agent by conducting tensile tests, and a linear relationship was proposed between the tensile failure stress and the crack opening distance. This failure stress ranged from 3.7 MPa for a crack open displacement (COD) of 50.8 μm (COD) to 1.2 MPa for a COD of 381 μm. Furthermore, based on the finite volume technique, a computational fluid dynamics model was established to investigate the internal features of the flow of healing agent, considering the contact properties between the self-healing agent and concrete surface.

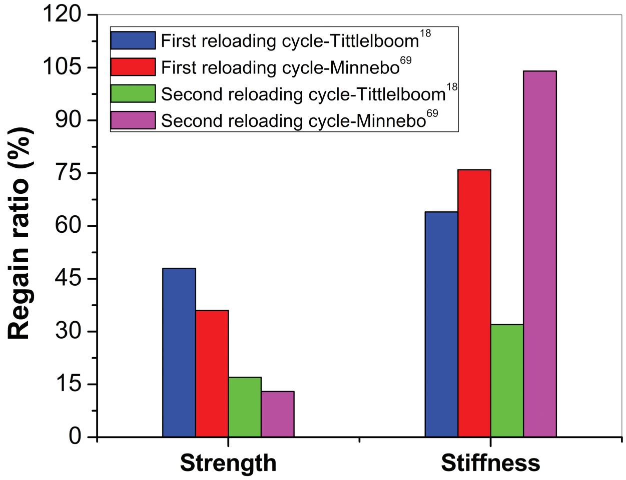

FIGURE 3 Comparison on self-healing efficiency of polyurethane-based healing agents[18,69]

Although various studies have been conducted to improve the self-healing efficiency of PU-based healing agents, there is still no agreement on the specific assessment criteria for self-healing efficiency. More controversially, for the encapsulated PU-based healing agent in large-scale concrete beams, four-point bending results are even variable due to the open crack interfaces and inadequate amount of healing agent.[68] The regaining ability of mechanical properties of PU-based healing agent are shown in Figure 3. The variation of the regained mechanical strength is much less than that of regained stiffness, whether in the first or second reloading stages. However, the efficiency of regained stiffness studied Minnebo et al.[69] was 104% during the second reloading, which was significantly higher than that of 50% by Tittelboom et al.[21] Further comparison convincingly reveals that the crack propagation behavior, the kinetics of healing agent in discrete crack surface, and other factors may also contribute to this significant deviation, except the capsule material or healing agent.

3 | DIFFERENT ENCAPSULATION TECHNIQUES

As the container of self-healing agents, the embedded capsules should be rupture as soon as cracks appear, and release healing agents immediately. Therefore, high brittleness is necessary for the qualified capsules. However, high survival ratio (robustness), low porosity, no interaction with healing agents, excellent bonding strength, comparable mechanical properties, and resistance to humid conditions are also ideal properties for capsules in self-healing cementitious material. On the other hand, geometry parameters of the capsule also affect the self-healing performances of concrete. Among all the developed encapsulating materials, glass and ceramic have gained the highest preference in laboratory-scale tests, despite the current little suitability in concrete repairing application due to the low survival ratio during concrete mixing. Therefore, it is necessary to pay attention to the advancements of glass and ceramic capsules for self-healing and summarize these current status of these novel capsule material.

3.1 | Glass and ceramic capsules

Li et al.[40] reported that the fitness of glass capillaries as carriers could be evaluated through in-situ observation of the breakage of glass capillaries and the release of healing agents. Similarly, Tittelboom et al.[47] distinguished the high enregy events caused by beakage of ceramic tubes using acoustic emission analysis, justyfing that the PU healing agent can be autonomously released from ceramic tubes. However, they both suggested that the high cost and fragility of these kinds of tubes are challenges to promote practical application.[32,40] Therefore, low survivability of glass or ceramic capsules during mixing in the realistic concrete structure is the great obstacle for the self-healing concrete technology.[46,56,70] Two different methods were proposed to protect the brittle capsules from damage during mixing.[46] The first method is to wrap capsules with a cord, and then cover them with a small layer of mortar. The second one is to use cement paste bars (water-to-cement ratio of 0.4): first put a layer of cement paste into the molds, subsequently, lay capsules on it, then fill with cement paste and conduct vibration. After 1-day casting, the concrete specimen can be prepared. However, Tittelboom et al.[46,47] reported that capsules in the second method may be broken by the mixing process. However, by hand-mixing, both of the methods are suitable for self-healing. Similarly, Thao et al.[31] suggested that wrapping a 6.5 mm layer of cement mortar around the glass capsules before casting can restrict premature breakage. Latterly, they proposed that protection by spiral wire coated with cement mortar by thickness of 3.5 mm is also effective.[42] All these protection methods are useful for improving the survivability of capsules, but skilled workmanship is still required to realize these schemes.[71]

More detailed research results regarding glass and ceramic capsules are shown in Table 2. The internal diameter of tubes ranges from 0.8 to 4 mm. Technically, internal diameter of tube determines the capillary forces, which pushes the healing agents out of the capsules.[51] However, Tittelboom et al.[21] revealed that there are no significant differences when the internal diameter of glass tube varies from 2 to 3 mm. Wall thickness of capsules is an important factor for increasing survival ratio during mixing. On the other hand, thicker wall of capsules may delay the healing process.[71] According to existing studies, tubes with external and internal diameters of 3.35 and 3.00 mm are usually used for self-healing.[21,30,32] In terms of the length of tubes, Tittelboom et al.[46] embedded continuous tubes in length of 400 mm in concrete beams, and found that the broken capsules was unlikely to release any PU during the four-point bending test. The reason is that the attractive force inside the tube is larger than the capillary one that drags the healing agent into the cracks. Therefore, short capsules seem to be more suitable for concrete self-healing. Compared with spherical capsules, cylindrical tubes exhibit a high probability for crack to propagate through.[63] Moreover, the bond strength between spherical capsules and cement matrix should be stronger than that of capsules to make sure the cracks can propagate through the capsules.[72] In addition, compared to spherical capsule, cylindrical tube can provide enough bound strength for the healing agent to heal the cracks.[73]

The self-healing efficiency of glass tube and ceramic tube with the same internal volume but different internal diameters was compared by Tittelboom et al.[21,46] In terms of the regained mechnical properties, there is a slight difference between ceramic capsule and glass tube, as shown in Figure 4. It seems that the glass capsule is better in mechanical strength regaining, during both the first or the second reloading. However, there is a variation in stiffness recovery abilities for these two kinds of capsules. Besides, ceramic tube is likely better choice for capsules to avoid alkali-silica reaction (ASR). The uncertainty of self-healing efficency between these two kinds of capsules also appears in the water permeability results. When the single crack propagates perpendicular to the capsule in Test 4 as shown in Figure 5, glass tube is able to reduce the water permeability coefficient by a factor from 10^2 to 10^3, which is lower than that of ceramic tube with a factor from 10^3 to 10^4.[21] Nevertheless, in another test where the crack widths, crack number, and capsule orientations are random in tests from Test 1 to Test 3, it is difficult to compare the efficiency between glass and ceramic capsules.[46] The accuracy of water permeability test is one of the reasons for this variation, but the crack patterns may be responsible for this uncertainty. More released healing agents are qualitativly visualized by high resolution

TABLE 2 Different capsule materials for self-healing cementitious materials

Literatures	Internal diameter (mm)	External diameter (mm)	Length (mm)	Capsule shapes	Capsule materials	Healing agents	Cracks	Test results
Tittelboom et al.[21]	2.00	2.20	41.3	Cylinder	Glass	Polyurethane	Single	More healing agent was localized in tubes
	3.00	3.35	18.4	Cylinder	Glass			No difference with ceramic ones in mechanical test
	3.34	3.86	15.0	Cylinder	Ceramic			Performed better in water permeability test
Lark et al.[49]	0.8	—	100	Cylinder	Glass	Cyanoacrylate	Single	Too fragile to be embedded
	1.5	—	100	Cylinder	Glass			
	3.0	—	100	Cylinder	Glass			Suitable capillary force
Tittelboom et al.[30]	3.00	3.35	50.0	Cylinder	Glass	Polyurethane	Multiple	No improvement in water ingress measurements
Gardner et al.[51]	1.3	—	87.5	Cylinder	Glass	Cyanoacrylate	Single	Similar capillary rise time for healing agents
	2.0	—	58	Cylinder	Glass			
Tittelboom et al.[46]	3.00	—	60	Cylinder	Glass	Polyurethane	Multiple	
	3.00	—	400	Cylinder	Glass			Short capsules released enough healing agents
	3.00	—	60	Cylinder	Ceramic			No difference in self-healing efficiency
Maes et al.[63]	3.00	3.35	50	Cylinder	Glass	Polyurethane	Single	Higher possibility than spherical ones for encountering cracks
Lv and Chen[22]	1.00	—	—	Cylinder	—	—	Single	Hitting probability was relevant to aspect ratio of capsules
	1.00	—	—	Spherical	—	—		

X-ray computed tomography (HRXCT) in concrete with ceremic tube than with glass tube, which is due to the difference in surface tension. The difference in amount of released healing agent may be the reason for the slight deviation presented in Figure 5.

3.2 | Polymer-based capsules

Capsule material development mainly aims at improving the survival ratio during concrete mixing by introducing different types of polymers,[74,75] which is summarized in Table 3. After being heated, the polymer capsules shift from a brittle state to a rubbery state prior to mixing with other components, thus the survivability of capsules considerably increases. Hilloulin et al.[74] investigated three kinds of polymers which were brittle at room temperature with a relatively low glass transition temperature to investigate the suitable materials for capsules. The capsule fabricated from MMA is the only materials which can release healing agent promptly among all the lactic acid, polystyrene, and MMA. Similarly, a specific type of poly(methyl methacrylate) (PMMA) is considered to be suitable for capsule materials.[75] This kind of capsule is able to rupture at average crack sizes between 69 and 128 μm, when the shell thickness varies from 0.3 to 0.7 mm. Moreover, the PMMA tube shows promising results in terms of the compatibility with various self-healing agents. Mostavi et al.[53] developed a double-walled PU/urea-formaldehyde (PU/UF) microcapsule filled with sodium silicate, and revealed that low pH value, high agitation rate, and high curing temperature can improve the formation of capsule shells. Optical microscope results confirmed that the self-healing microcapsule contains phenol-formaldehyde resin as shell and dicyclopentadiene as healing agent, respectively.[76]

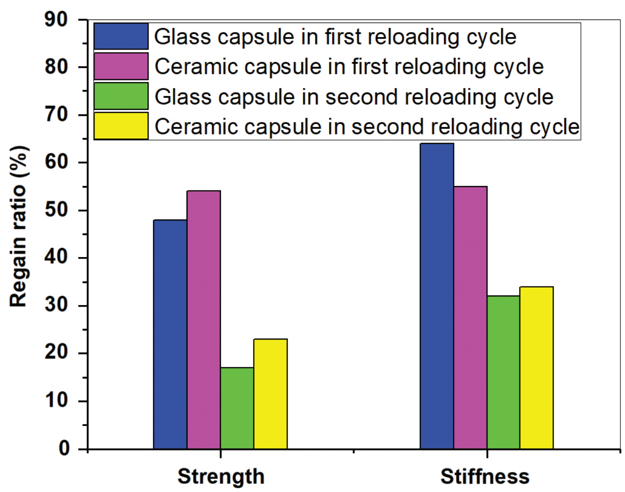

FIGURE 4 Comparison on self-healing efficiencies between the class tubes and ceramic tubes after reloading[16]

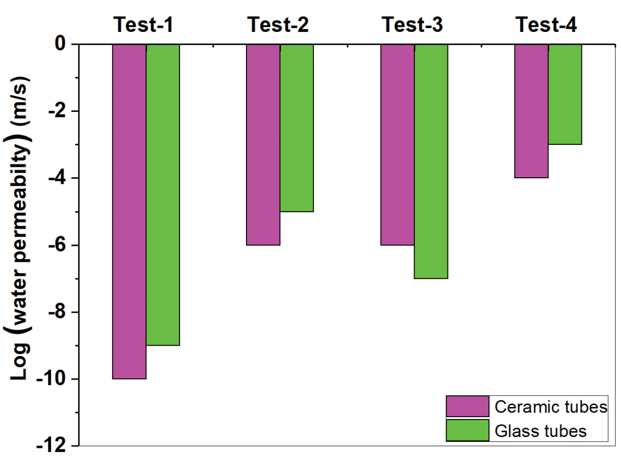

FIGURE 5 Comparison on self-healing efficiencies between class tube and ceramic tube in term of water permeability[16,40]

TABLE 3 Comparison on polymeric capsules for self-healing concrete

Literature	Capsule materials	Healing agents	Test results	Disadvantages
Hilloulin et al.[74]	Lactic acid Polystyrene Methyl methacrylate	Polyurethane-based	Only methyl methacrylate capsule released healing agent	Heating was required
Van Belleghem et al.[85]	Methyl methacrylate Polystyrene Polylactic acid	— — —	Only methyl methacrylate capsule ruptured	Large elongation
Lv et al.[76]	Phenol–formaldehyde	Dicyclopentadiene	No interference with matrix	Low rupture probability
Yang et al.[57]	Silica gel	Methyl methacrylate	Compatible	Low rupture probability
Beglarigale et al.[109]	Sodium silicate solution	—	Sufficient interfacial bond strength	Complex synthesis process
Mostavi et al.[53]	Urea-formaldehyde	Sodium silicate	PH and temperature affect manufacturing	Complex manufacturing process
Ni et al.[77]	Urea-formaldehyde resin	Epoxy	No loss in mechanical strength	—
Minnebo et al.[69]	Inorganic phosphate cement	Two-component polyurethane	Less strain concentration	High cost

Note: PH = Power of hydrogen.

When capsules are embedded into the concrete, a weak zone may be created due to the strength difference between these two types of materials. The volume of released healing agent increases with the number of capsules due to high probabilities of the capsules breakage by cracks. However, the mechanical strength of concrete structure may be reduced in turn, therefore the adjustment of capsule dosage is required for better self-healing performance.[71] Theoretical dosage of capsules required for crack repairing was developed by Lv et al.[22] They suggested that the volume fraction of capsules incorporated in cement matrix is opposite to the number of crack size. Meanwhile, probability of cylindrical capsules broken by cracks is not always higher than that of spherical capsules, and the healing efficiency may be also relevant to aspect ratio of capsules. As the current studies mainly concentrate on the single controlled crack and embedded capsules, the balance between the number of capsules and healing efficiency is still challenge. The polymeric capsule may be able to minimize or even eliminate the mechanical strength deterioration.[74] When UF resin spherical microcapsules filled with epoxy were incorporated, no loss in mechanical strength was found by Ni et al.[77] Besides, inorganic phosphate cement (IPC) tube seems a novel design for capsules.[69] Compared with traditional ceramic tubes, the IPC capsule is unlikely to decrease the mechanical properties of concrete beams, and more importantly, can reduce local strain concentration.

4 | ENCAPSULATION-BASED SELF-HEALING CONCRETE

4.1 | Initiation of self-healing action

The self-healing efficiency of encapsulation-based self-healing concrete is primarily evaluated from initiation of self-healing process by creation of cracks to assessment of self-healing performances.[78] The related test approaches are classified in Table 4, including initiation of self-healing reaction,[21] evaluation of self-healing efficiency,[30] and monitoring of healing process.[47] Typically, three-point bending test is widely adopted to create the preoriented crack in prism samples, and simultaneously monitor the self-healing process.[21,32,66] Three-point bending test is usually conducted with linear variable differential transformer (LVDT), which is attached at the bottom of the specimen to measure the crack width. Commonly, in order to control the crack propagation, a notch of the specimen was prepared perpendicular to the preplaced capsules.[66] Multiple cracks in the beam are usually created by four-point bending tests.[30,46,79] In this system, the width of each individual crack is approximately calculated though dividing the LVDT value by the total amount of cracks, when the elongation of the specimen is ignored.[30]

TABLE 4 Classification of test methods for self-healing concrete

Items	Mechanical tests	References
Initiation of self-healing action	Three point bending test	21,32,66
	Four-point bending test	30,46,79
	Splitting test	21
Evaluation of self-healing efficiency	Three point bending test	21,32,66
	Four point bending test	30,46,79
	Tensile test	21,110
	Water permeability	21,30,32,46,47
	Capillary water uptake	58,84,85
	Digital image correlation	66
	Micrograph	66
	X-ray computed tomography	46
	Florescent microscopy	30
	Neutron radiography	58
	Infrared analysis	26,61
Monitoring of self-healing process	Acoustic emission analysis	46,47,66
	Piezoelectric transducers	51,61,62

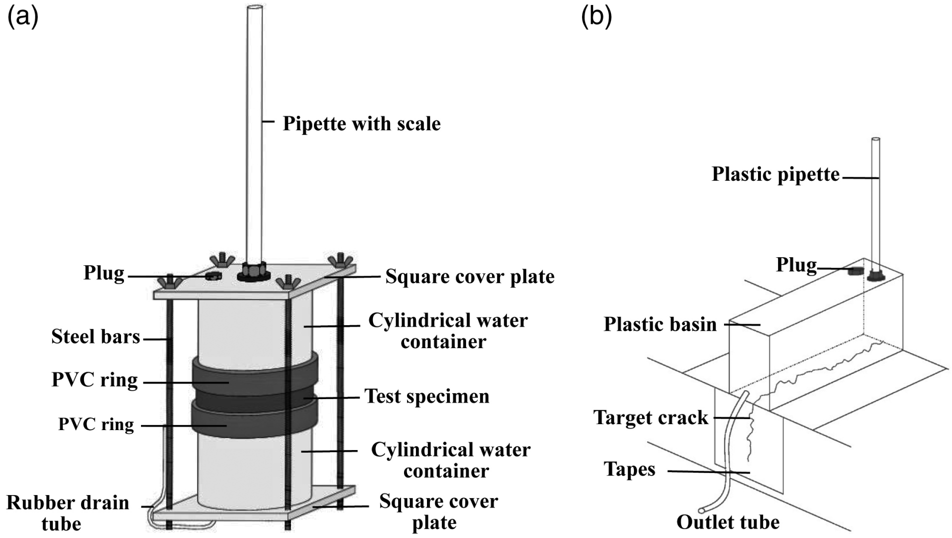

FIGURE 6 Regained mechanical properties of self-healed concrete after different reloading cycles[16]: (a) single preoriented crack; (b) multiple free cracks

4.2 | Evaluation of self-healing efficiency

4.2.1 | Durability regaining

Promoting durability recovery is the primary goal for self-healing concrete, which determines indication for self-healing efficiency assessment. Fluid permeability is a direct indication for concrete durability to evaluate the service life of concrete infrastructure.[3,80–83] Therefore, permeability coefficient test is the most prevailing method to assess the durability of self-healing concrete. Tittelboom et al.[21] provided complete details for the setup and procedure of water permeability test. The water permeability coefficient is determined by recording the decrease of water column with duration, as show in Figure 6a, and is quantitatively calculated using Darcy's law. This approach has been extensively applied to measure healing efficiency of single preoriented crack, and proves that the encapsulation strategy is very effective in durability recovery. Tittelboom et al.[21] measured decreases in water permeabity of concrete with cured cracks, and the results varied form 10^2 to 10^4 for concrete beam containg different capsules. However, for the large-scale test with multiple cracks in Figure 6b, it is hard to evaluate the healing efficiency, and the water permeability of each crack is difficult to be measured as well.[30,46]

Compared to the permeability test, capillary absorption coefficient seems more accurate to evaluate the durability of crack-healed concrete in the unsaturated service.[58,84,85] Conventionally, gravimetrical method has been extensively utilized to calculate the quantitative capillary absorption coefficient in a simple way, but is unable to display the water spatial distribution.[32] Therefore, X-ray computed tomography and neutron radiography techniques have been applied to obtain details of moisture distribution inside the discrete cracks. Tittelboom et al.[58] first visualized the water absorption of different specimens by neutron radiography and the water distribution in the healed crack surfaces, then analyzed the effect of viscosity on self-healing performance.[84] Although the resolution and accuracy are very effective, the high cost of neutron radiography test restricts its further prevalence. Therefore, it might be economical to assess the durability of crack-healed concrete by X-ray radiography technique with affordable cost.[85]

In addition to water permeability or capillary water absorption investigations, few studies focused on chloride penetration of encapsulation-based self-healing concrete.[86] Maes et al.[63] studied the self-healing performance through acid-soluble chloride extraction test and found that encapsulated PU is able to prevent chloride diffusion from crack with width from 100 to 300 μm. However, as for large cracks, the insufficient healing reaction causes high chloride permeability, due to the lower capillary force between crack surfaces than that in tubes.[49] In addition, Yang et al.[57] concentrated on gas permeability coefficient, and found that substantial reduction of around 50.2% for self-healing cement mortar by oil core/silica gel shell microcapsules.

4.2.2 | Mechanical properties regaining

When self-healing process initiates and the healing agent completely hardens, the self-healing concrete is reloaded to measure the strength, stiffness, and toughness based on the stress–strain relations or force-displacement curves by three- or four-point bending tests. Tittelboom et al.[21] evaluated the self-healing efficiency of encapsulation-based self-healing concrete by comparing loading curves of untreated, manually cured and autonomously healing methods. The peak load (F_c) and the slope of the force–displacement curve are corresponding to strength and stiffness indications, respectively, the regaining efficiency of mechanical properties for autonomous healing concrete is demonstrated in Figure 7. Feiteira et al.[32] conducted three-point bending test to track the force–displacement response of cracking and crack widening cycles for the healed and nonhealed cement mortar with encapsulated polymer precursors. Due to low stiffness

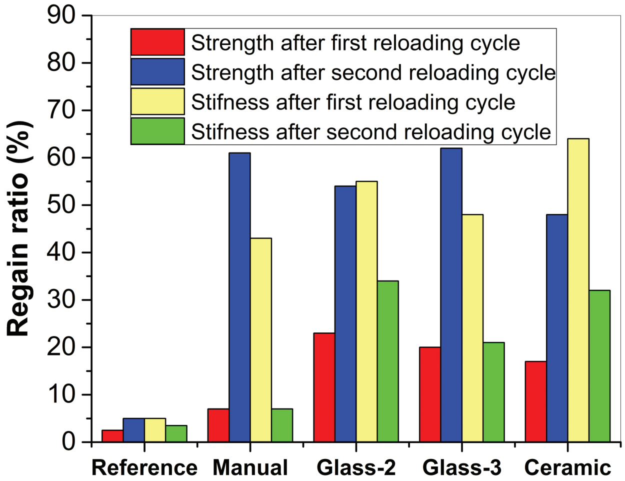

FIGURE 7 Water permeability test setup for self-healing cementitious materials[16,24]

of polymers used, they only obtained regaining of 30% mechanical stiffness for crack month with thickness up to 20 μm. Similarly, during the repeated loading cycles, Thao et al.[31] found an approximately 30% increase in strength with respect to the initial strength. However, in another mechanical test,[47] over 80% of the original strength and stiffness were regained by encapsulation-based self-healing method. Thus, with the same healing agents and capsules, the regaining capacity of mechanical strength may be significantly affected by the crack patterns. For example, as for self-healing concrete with multiple cracks, the four-point bending test is unable to obtain desirable results on regaining the regaining capacity of mechanical strength.[30] Moreover, some unexpected mechanical recovery results were also yielded for concrete beam subjected to four-point bending load by Karaiskos et al.[79]

4.2.3 | Self-healing performance visualization

Digital image correlation (DIC), micrograph, HRXCT, and florescent microscopy are predominant methods for visualizing self-healing mechanism and process, such as crack width, capsules location, and healing agent releasing. Tittelboom et al.[30] adopted all the abovementioned techniques to visualize and to evaluate the self-healing behavior of PU-based healing agent in large-scale test. The different results are summarized in Figure 8. DIC is usually applied to analyze the crack patterns on concrete surfaces.[87,88] According to the DIC results in Figure 8a, it is clear that a denser crack pattern occurred in the middle part of the concrete beam. Feiteira et al.[66] also used DIC to monitor the crack opening along the full height of the crack, and observed a more severe crack widening process in the case of rigid polymers as healing agent. X-ray tomography has been performed to obtain 3D visualization of internal part of concrete to investigate the location and status of capsules. From Figure 8b, the capsules are located at a depth of around 13 mm below the top surface of the specimen. It was found that the capsules are not completely emptied due to the insufficient capillary force, and the healing agent within the crack spreads discontinuously. The same phenomenon was confirmed using the micro-CT image analysis by Gilabert et al.[67] Based on the X-ray radiographs of the concrete beam slices with realistic cracks, Tittelboom et al.[46] concluded that whether a crack propagates through a capsule is based on coincidence, unless the cement paste bars act as weak zones between capsule and cement matrix and attract the crack initiation. Micrographic representation techniques are usually applied to quantitatively determine the width of crack and visualize the distribution of healing agent, as shown in Figure 8c. It shows that the beam with encapsulated PU exhibits the highest portion of crack sealing and the strongest ability to heal larger cracks with width of 189 μm. In addition, Feiteira et al.[32,66] visualized the detachments from cracks of encapsulate polymer by the microscope observation.

4.2.4 | Comparison on evaluation methods

The self-healing efficiency assessment methods from different studies are compared in Table 5. It is evident that the regained mechanical properties are the main indication for self-healing ability assessment, followed by water permeability, capillary absorption coefficient, and so on. More and more researchers combine these tests with some nondestructive testing techniques, such as DIC, X-ray computed tomography, neutron radiography, and acoustic emission (AE) analysis. The results show that few researchers have conducted all these kind of experiments to evaluate the self-healing efficiency, especially the high-cost neutron radiography. Tittelboom et al.[21,47,58] applied most of these methods to promote the application of self-healing technology for cementitious materials. Although Table 5 lists the limitation of each method, it is still difficult to choose the most reliable one, because of the ideal preoriented cracks far from the practical engineering ones, the complex behavior of cementitious materials, the low precision of experiments, and the high cost of test facilities.

Among all these approaches, the precision of water permeablity and capillary tests still need further improvement. Although assessment method, healing agent, capsules, and raw materials of the specimens are the same except the amount of cracks, the test results of experiments may be entirely contrary to the other ones. In Tittelboom et al.,[21] ceramic capsules performed better than glass capsules in water permeability tests, which is not consistent with the results of Tittelboom et al.[46] There are still contradictory arguments on whether the X-ray computed tomography is able to detect the crack-healing or not depending on its limited resolution. Compared with the low-cost ultrasonic pulse velocity detecting technology, more and more researchers prefer to combine AE analysis with DIC to monitor the crack formation and capsule status.[30,89] Electronic microscopy, environment electronic microscopy, and the rarely adopted

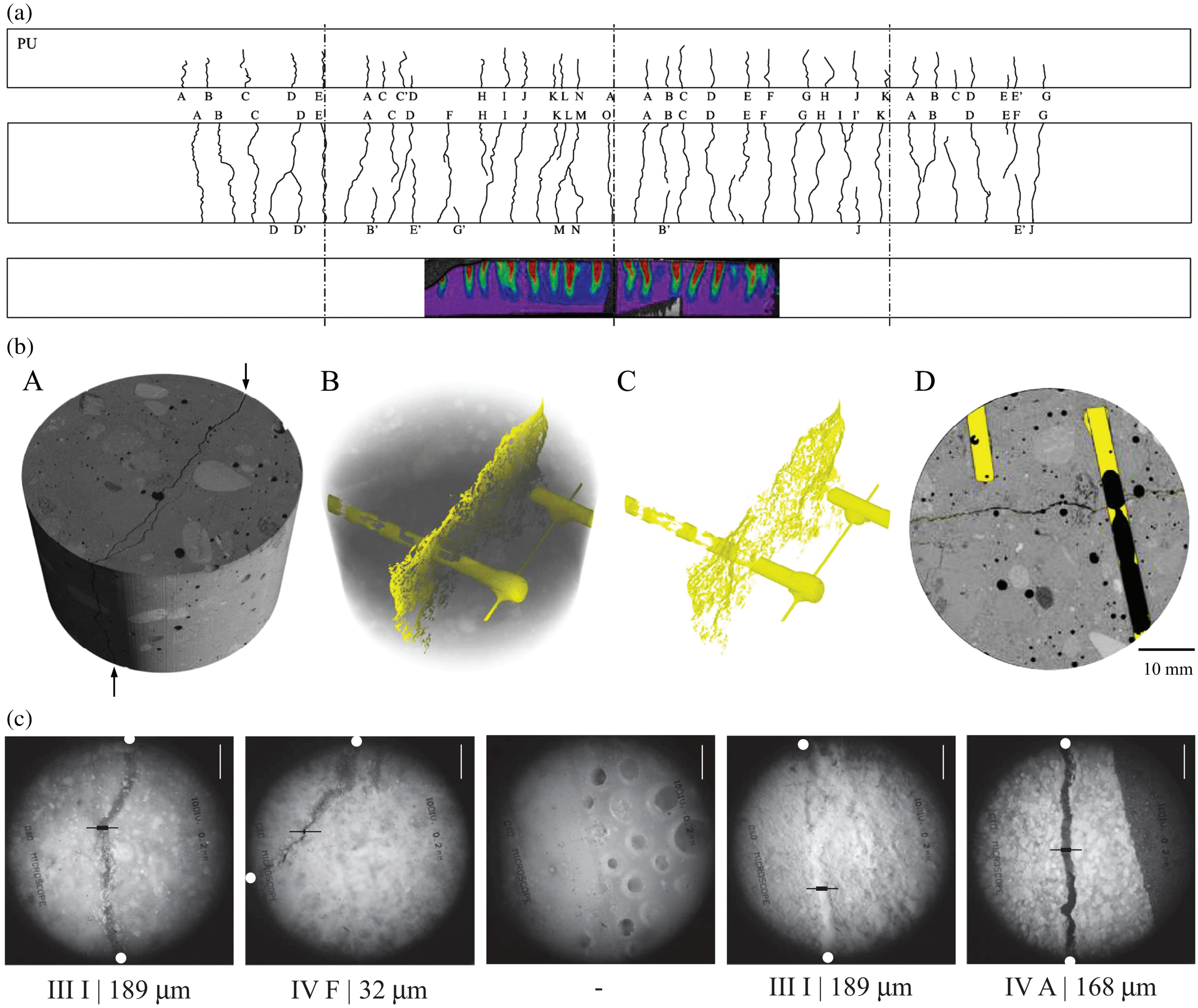

FIGURE 8 Visualization of self-healed cracks in concrete by different methods[24]: (a) digital image correlation; (b) X-ray tomography; (c) microscale graph

fluorescent microscopy are the most useful methods to conduct the optical investigation on self-healing behavior.

4.3 | Monitoring of self-healing process

AE analysis has been widely used to monitor the crack propagation and self-healing process in concrete, because high energetic release induced by tube breakage can be recognized during both cracks propagation and capsule breakage.[46,47,66,90] Different phases of crack formation were distinguished during the reloading stage for the healed concrete using AE analysis by Tittelboom et al.,[47] which indicates the multiply healing action of encapsulation-based self-healing techniques. On the other hand, it was found that when the acoustic emission was applied to detect detachment failure of healing agent form cracks surface during reloading of self-healed specimens, only the healing agent detachment of rigid polymer foams with elastic modulus of 22 MPa caused distinguishing high-energy events.[66] Besides, Dumoulin et al.[91] embedded piezoelectric transducers in concrete structure to detect the crack initiation phase, and monitored the crack propagation over the entire duration of three-point bending test. Karaiskos et al.[79] also applied piezoelectric transducers to record the multiple crack formation and propagation of concrete containing PU-based capsules in four-point bending test. Compared with AE technology, the piezoelectric transducer depends on monitoring the ultrasonic pulse velocity, and is more suitable for online operation, due to their low cost, small size, and broad frequency band.[47,92–96]

4.4 | Numerical investigations

Compared with experimental methods, numerical simulation seems more convenient and low-cost, although the accuracy is sacrificed through idealizing the real self-healing behavior and the complex properties of cementitious materials.[97–99] Nevertheless, numerical analysis is a valuable technique to investigate self-healing mechanics with parametric analysis, if there are

TABLE 5 Evaluation of self-healing efficiency for cementitious materials

Mechanical loads	Properties	Limitations	References
Three/four bending test	Initiation of self-healing Evaluation of mechanical strength	Strongly affected by the cracks states; influence of empty capsules could not be excluded	16,21,30,31,33,40,44,46–48,56,74,79,111,112
Tensile tests	Calculation the mechanical property	Low accuracy	32,67,74
Dynamic mechanical analysis	Storage modulus	Significant alteration under different test temperature	44,74
Water permeability/ capillary	Durability assessment	Precision and efficiency depending on the cracks states	21,30,32,46,51,56,84,85,111
Chloride permeability	Chloride penetration measurement	Indirectly realized	4,63,71
X-ray computed tomography	Leakage of healing agent and capsule states	Limited resolution	21,26,30,85
Neutron radiography	Water uptake investigation	High cost	58,84
Microscopy/SEM/ESM	Healing results measurement and failure mode analysis	Unsuitable for fragile and large specimens	15,16,26,30,40,44,61,66,113
Infrared analysis	Composition determination of samples	Moisture decreasing the accuracy; unavailable for minor deposition	26,61
Digital image correlation	Crack patterns and strain distribution	Precision relies the camera and calibration analysis	30,40,66
Fluorescent microscopy	Depth of the healed cracks	The sample must be fluorescent	30

Notes: ESM = Electron Scanning Microscopy; SEM = Scanning Electron Microscopy.

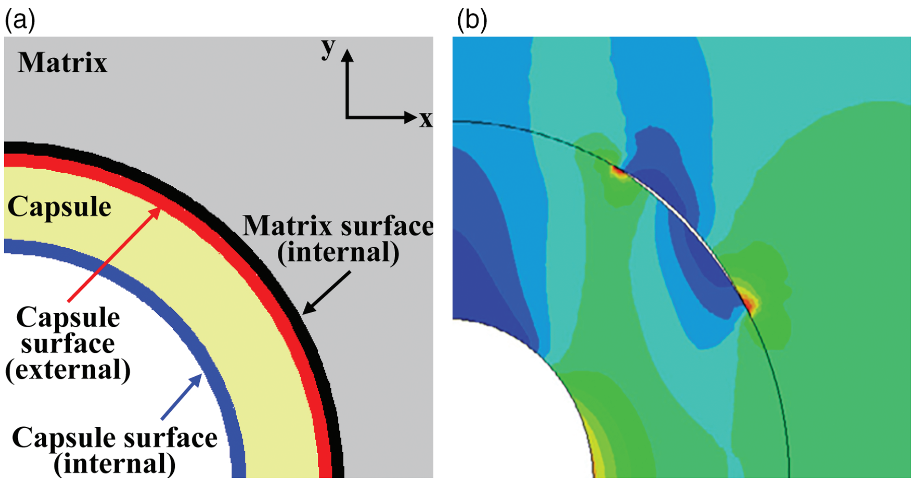

FIGURE 9 Two-dimensional symmetry quarter plane strain model for stress concentration analysis on self-healing concrete[18]: (a) numerical model; (b) interface debonding

improvements in assumptions of the nonuniform and multiaxial load conditions and consideration of the inconsistency between the capsules and cement matrix.[100–105] Associating with self-healing process, the simulation researches mainly involve initiation of self-healing action, flow of healing agent and required quantity of capsules and healing agent.

Gilabert et al.[106] combined eXtended Finite Element Method (XFEM) elements and cohesive surface (CS) techniqus to predict crack propagation and brekage of the capsules. They simulated the response of a beam with encapsulated systems in a three-point bending test, concluding that when using capsules with t/R less than 0.12, a minimum interface strength of 2.0 MPa has to be ensured to break the capsules and liberate the healing agent. In Figure 9, a detailed two-dimensional symmetry quarter plane strain model was established to analyze stress concentration and bonding strength in encapsulation-based self-healing materials.[23] The numerical simulation results demonstrate that the initiation of deboning is dominated by strength ratio, geometric ratio, and elastic modulus ratio. However, the specific value of bonding strength was not determined in this art of paper. Therefore, latterly, an axisymmetric 3D finite model was built to assess strength and fracture toughness of glass-concrete interface.[33] The verified interface bonding strength is 0.96 ± 0.09 N/mm^2 calculated by cohesive zone model, and the interfacial energy is 10.62 ± 0.02 mJ/m^2 obtained by virtual crack closure technique.

Concerning the flow of healing agent and efficiency of self-healing, a computational fluid model called "inter foam" was proposed to analyze the relationship between spatial distribution of healing agent and the crack width.[67,107] The computational domain includes the interior of capsules and

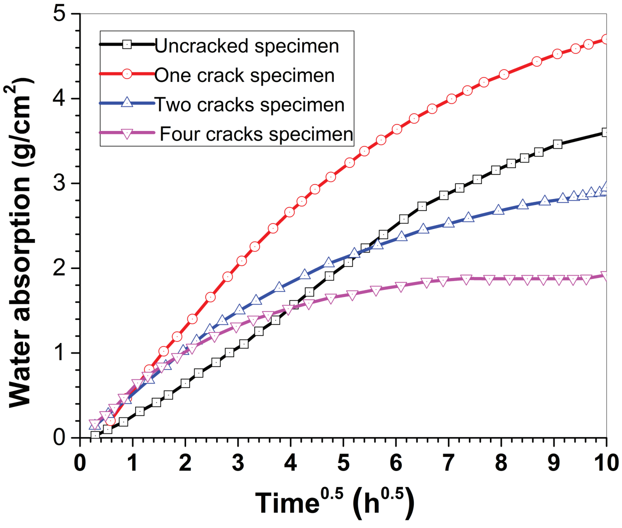

FIGURE 10 Comparison on cumulative water absorption per unit of surface area exposed to water[78]

the space between two crack surfaces. Simulation results revealed that for the case of COD of 300 μm, the released resin was located in half way to the bottom of crack surfaces after 30 s. On the other hand, in terms of the narrow cracks with COD of 100 μm, a stationary state was reached only after 15 s, considering the contact properties between the healing agent and the concrete surface. Similarly, Van Belleghem et al.,[85] analyzed the water absorption in cracked cement mortar using Richard's equation and vaporization process natural boundary condition. From Figure 10, the finite element analysis results revealed that water absorption of per unit of surface area is less, when the concrete prisms contain more than one cracks. Besides, Gardner et al.[51] simulated the capillary flow of cyanoacrylate healing agent in small glass capillaries, and predicted the increase in capillary flow, which suggests that the influence of wall slip on the capillary can be ignored. According to the targeted healing level, a philosophy was recommend by Lv et al.[22] to determine the required dosage and volume fraction of capsules to fulfill the healing expectations. Meanwhile, two mathematical models were developed to calculate the probability of crack hitting a capsule. The built functions allow analyzing the efficency of a self-healing material with accounts of crack length, capsule size, and mean intercapsule distance.[20]

5 | CONCLUSION AND SUGGESTION

Encapsulation-based self-healing technology has been widely used as an effective method for crack healing of concrete. Healing agents and encapsulation techniques are the key factors for the encapsulation-based self-healing technology. There are various experimental and numerical studies to evaluate the efficiency of self-healing method. The related conclusions and suggestions are drawn as follows:

1. Among all available self-healing agents, PU-based polymer is the most promising, with high flexibility and comparatively shorter curing time and self-healing process without water. However, further modifications are still needed for stress concentration around the interface, detachment of hardened self-healing agent, incomplete mixing, and premature reaction.
2. Glass and ceramic are the most widely used capsule materials, especially in the laboratory scale studies. However, low survivability of capsules during mixing is a challenge for improving self-healing concrete techniques. The polymer capsule exhibits a promising ability in improving the survival ratio and minimizing mechanical strength loss, but complex heating mixing process, low chances of breakage, and high elongation at rupture may compensate these advantages.
3. The absence of standard self-healing efficiency indicators and evaluation criteria make it difficult to determine the most efficient and reliable efficiency assessment method for self-healing behavior. The mechanical tests and permeability tests are widely applied as basic methods to investigate the efficiency of encapsulation-based self-healing concrete. Nondestructive testing techniques such as DIC, X-ray computed tomography, neutron radiography, acoustic emission analysis, and numerical simulation can together provide better understanding of self-healing behavior.
4. There is still a long way to reach a consistent conclusion on suitable viscosity, fitted elastic modules, strain capacity, and bonding strength of PU-based polymers healing agent. Except the capsule materials and healing agents, the crack propagation behavior of encapsulation-based self-healing concrete, kinetics of healing agent in discrete crack surfaces, and other comprehensive factors are all worth further investigations.

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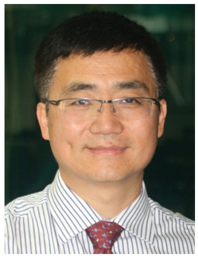

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