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A state-of-the-art review

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The use of additive manufacturing in self-healing cementitious materials: A state-of-the-art review

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ABSTRACT

This paper presents a state-of-the-art review on the application of additive manufacturing (AM) in self-healing cementitious materials. AM has been utilized in self-healing cementitious materials in three ways: (1) concrete with 3D-printed capsules/vasculatures; (2) 3D concrete printing (3DCP) with fibers or supplementary cementitious materials (SCMs); and (3) a combination of (1) and (2). 3D-printed capsules/vascular systems are the most extensively investigated, which are capable of housing larger volumes of healing agents. However, due to the dimension restraints of printers, most of the printed vasculatures/capsules are in small scale, making them difficult for upscaling. Meanwhile, 3DCP shows great potential to lower the environmental footprint of concrete construction. Incorporation of fibers and SCMs helps improve the autogenous healing performance of 3DCP. Besides, 3D-printed concrete with hollow channels as the vasculature could further improve the autonomous healing and scalability of self-healing cementitious materials. Finally, possible directions for future research are discussed.

1. Introduction

In nature, the survival of living organisms is facilitated by their ability to rapidly heal damage such as wounds or fractures. The self-healing capacity of organisms inspired the development of self-healing systems in polymer-based composites and more recently in cementitious materials (Zhang and Li, 2016; Hager et al., 2010). In the case of cement-based infrastructure and construction materials, self-healing could effectively lower the maintenance and repair costs. Based on a recent research (Nguyen et al., 2023), there are five main strategies for realizing self-healing in cementitious materials: (1) autogenous self-healing due to chemical causes: (a) *carbonation* and (b) *crystallization (such as continued hydration)*; (2) autonomous self-healing by (c) *biomineralization*, (d) *polymer-cement composites*, and (e) *fibers*. Autonomous self-healing can, based on the implementation approaches, be further classified into capsule based self-healing and vascular self-healing.

1.1. Self-healing cementitious composites

1.1.1. Autogenous self-healing in cement-based materials

Early approaches to self-healing of concrete mainly involved the stimulation of autogenous healing. In 1836, the French Academy of Science was the first to take notice of the autogenous self-healing phenomenon present in cement-based composites (Slate and F 0). Little progress has been made until last century when the mechanisms of autogenous self-healing were systematically investigated. Fig. 1 shows the possible causes of autogenous self-healing of cementitious materials in water. Compared with the physical and mechanical causes, chemical causes are more significant and easier to control. However, the crack sealing capacity induced by the autogenous self-healing is low: healable cracks range are in the size range of 10–100 μm (Belie et al., 2018).

To promote continuing hydration or carbonation of the dissolved $Ca(OH)_2$, a concrete mix can be designed or modified by directly incorporating mineral or crystalline admixtures to stimulate autogenous self-healing (Huang et al., 2014, 2016; Van Tittelboom and De Belie, 2013; Li et al., 2018; Ferrara et al., 2014, 2016). Furthermore, the presence of water is another significant stimulator for autogenous self-healing. Superabsorbent polymers (SAP) have been used to induce the

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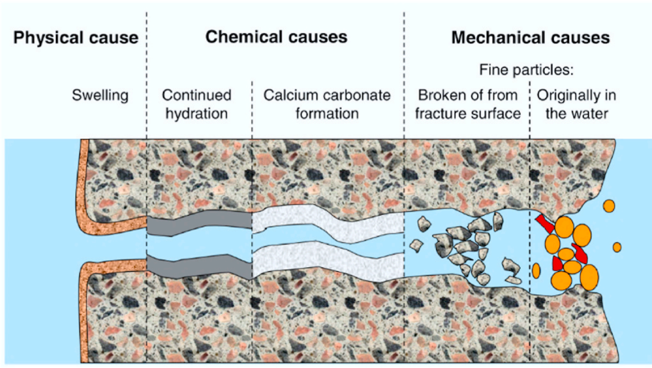


Fig. 1. Mechanisms of autogenous self-healing (Rooij et al., 2011).

self-healing of cementitious materials (Belie et al., 2018; Rodríguez et al., 2019). The swelling of SAP initially rapidly seals the crack while later providing water for continued hydration which may permanently seal the crack (Mignon et al., 2017). However, the maximum healable crack width by the simulated autogenous healing is roughly less than 200 μm and preferably lower than 100 μm (Snoeck et al., 2014; Ferrara et al., 2018).

1.1.2. Autonomous self-healing in cement-based materials

1.1.2.1. Capsule based self-healing cementitious materials. Compared with directly incorporating engineering additions in the cementitious matrix, encapsulation of healing agents is a more desired approach for delivering healing agents to the cracked region, enabling on-situ repair (Belie et al., 2018). This self-healing process is activated by capsule breakage, caused by e.g., cracking or damage. The released healing agents flow to the cracked area and thereby sealing the cracks. There are myriad healing agents which could be encapsulated for self-healing. Except for mineral or crystalline admixtures, polymeric healing agents can also be encapsulated to heal the cracks (through polymer-cement composites) (Dry, 2000; Kanellopoulos et al., 2017; Xue et al., 2019; Hu et al., 2018; Dong et al., 2017). An example of capsule based self-healing concrete is shown in Fig. 2.

Furthermore, microorganisms can be encapsulated to form self-healing bio-concrete in which calcium carbonate is a product of the biological processes which can seal cracks through biomineralization. The encapsulated bacteria are designed to be dormant until cracks form in the concrete. Due to the highly alkaline environment of the cementitious matrix, the selection of bacteria and capsule materials are of great importance (Wang et al; Tziviloglou et al., 2017; Tziviloglou et al., 2016). Several factors (i.e., curing environment and crack width) affect the precipitation efficiency and the healing effectiveness. For capsule based self-healing concretes, there are two main limitations: (1) the limited amount of healing agent in a single capsule does not allow for multiple healing processes in case of re-opening of cracks (Lee et al., 2018); (2) the random and uncontrolled distribution of capsules resulting from the concrete mixing process. The microcapsules are too small to be positioned in particular places.

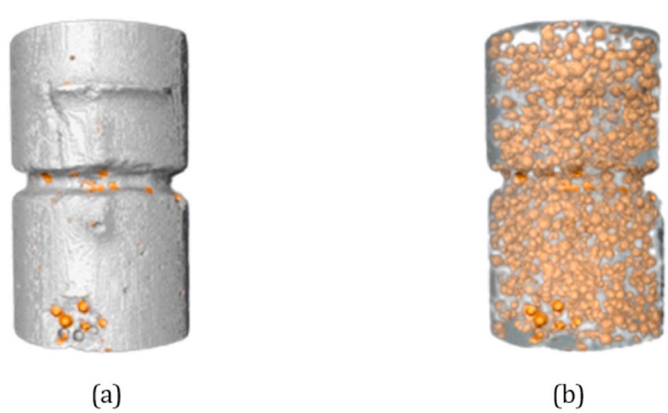


Fig. 2. Capsule based self-healing concretes (Lv et al., 2017) (a) Paste cylinder; (b) Microcapsules in cementitious matrix.

1.1.2.2. Vascular self-healing cementitious materials. Inspired by the vasculature in mammals or leaf venation of plants (Fig. 3a), incorporating vascular systems in cementitious matrix enables transporting healing agents to the cracked region (Fig. 3b). Compared with capsule based self-healing concretes, one of the main advantages of vascular self-healing concrete is the ongoing supply of healing agents, facilitating healing of wider cracks and repetitive healing process (Hamilton et al., 2010). Furthermore, the probability of triggering the healing process is larger than that of capsule based counterpart since the vasculature has larger probability to be hit by the crack. Similar to the capsule materials, the vascular material should be strong enough to survive concrete mixing and casting, but brittle enough to fracture when cracks in concrete occur (Sun et al., 2011). However, restricted by the brittle behaviour of vascular materials, the geometry of vascular systems in early researches used to be relatively simple mostly one-dimensional (tubes) or planar, especially when 3D printing is not involved (Joseph et al., 2010; Van Tittelboom et al., 2011; Shields et al., 2021a).

1.2. Additive manufacturing (AM)

Additive manufacturing (AM, also known as 3D printing) benefits the development of engineered materials due to its freedom of design and automation. There are six main methods of additive manufacturing, i.e., fused deposition modelling (FDM), powder bed fusion, inkjet printing and contour crafting, stereolithography (SLA), direct energy deposition (DED) and laminated object manufacturing (LOM) (Ngo et al., 2018).

1.2.1. AM in self-healing polymeric materials

For self-healing materials, vasculature with complicated geometry can be additively manufactured and embedded in the host matrix to increase the healing efficiency. Prior to cement-based materials, additive manufacturing has been widely used in polymers for self-healing. In the work of Toohey et al. (2007), self-healing polymers with microvascular networks were put forward. The 3D microchannel network was directly written with a fugitive ink, consisting of 60 wt% petroleum jelly and 40 wt% microcrystalline wax. After the completion of vascular fabrication, the heat treatment of the substrate, along with the application of a gentle vacuum, serves to eliminate the fugitive ink. For the self-healing materials with microvascular networks, 7 healing cycles are performed with a peak recovery of 70%. To accelerate the healing process and increase the healing performance, ternary interpenetrating microvascular networks were sequentially printed in epoxy substrate where two of the networks are for epoxy resin and hardener while the third one is employed for thermal control (Hansen et al., 2011). Similarly, a printed microvascular system with the double cleavage drilled compression (DCDC) fracture sample geometry was utilized to transport healing agents to the cracked region (Hamilton et al., 2010).

1.2.2. 3D printing of concrete (3DCP)

Except for being free of formwork, the design flexibility of 3D concrete printing (3DCP) (together with structural optimization) may enable reducing the waste of construction materials. Therefore, 3DCP shows great potential to lower the environmental footprint of concrete construction (Muthukrishnan et al., 2021; Bhattacherjee et al., 2021; Chen et al., 2021). Considering that printing cementitious ink plays a key role in the printing process, a lot of research focused on the development of printing materials, which are required to hold good

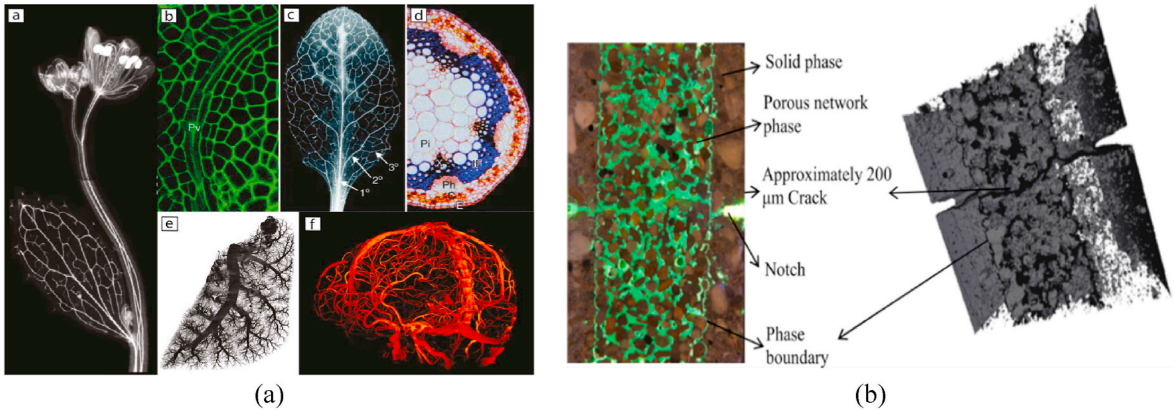


Fig. 3. (a) Vasculatures in nature (Qamar et al., 2019); (b) Mimicking bone healing in concrete (Sangadji and Schlangen, 2012).

pumpability and buildability (Muthukrishnan et al., 2021; Hou et al., 2021). Compared with conventional cementitious materials (for casting), superplasticizer (SP) and viscosity modifying admixture (VMA) are indispensable ingredients for 3D printing cement-based materials (Chen et al., 2019, 2023). Furthermore, to mitigate the conflict between pumping and deposition, accelerator could be sometimes injected close to the nozzle to develop the better buildability of the deposited structures by using a dynamic mixer (Lloret-Fritschi et al., 2020) or a static mixer (Tao et al., 2022).

It should be noted that coarse aggregate is usually absent in the 3D printable cementitious materials to ensure good extrudability. As a result, the 3D printable cementitious materials tend to be even more brittle than conventional concretes. To alleviate this issue, fibres have been added in printable mixtures (van Overmeir et al., 2022). As mentioned above, one way of promoting self-healing in cement-based materials is through use of fibres which may inhibit crack propagation and promote autogenous self-healing. Except for promoting self-healing of 3D printing concrete by optimizing mix design, another possibility is to tailor the structure of the printed concrete. For example, architectured hardened cement pastes fabricated with 3DCP are proven to have an enhanced fracture and damage tolerance compared with the cast elements from the same materials (Moini et al., 2018). This could also inspire the application of additive manufacturing in self-healing cementitious materials.

1.3. Strategies of using AM in self-healing cementitious materials

The possible strategies of using additive manufacturing (AM) in self-healing cementitious materials are shown in Fig. 4. The fabrication and characterization of capsule/vasculature for self-healing cementitious materials through additive manufacturing (mainly through FDM) are reviewed in section 2. Subsequently, 3D printing of self-healing concrete including autogenous healing and autonomous healing in 3DCP is discussed (section 3). Finally, the future challenges for further applying additive manufacturing in self-healing cementitious materials are discussed.

2. Self-healing concrete with 3D-printed macro-capsules/vascular system

2.1. 3D printing of macro-capsules

As mentioned in section 1.1.2, the healing performance of capsule based self-healing cementitious materials is restricted by the limited

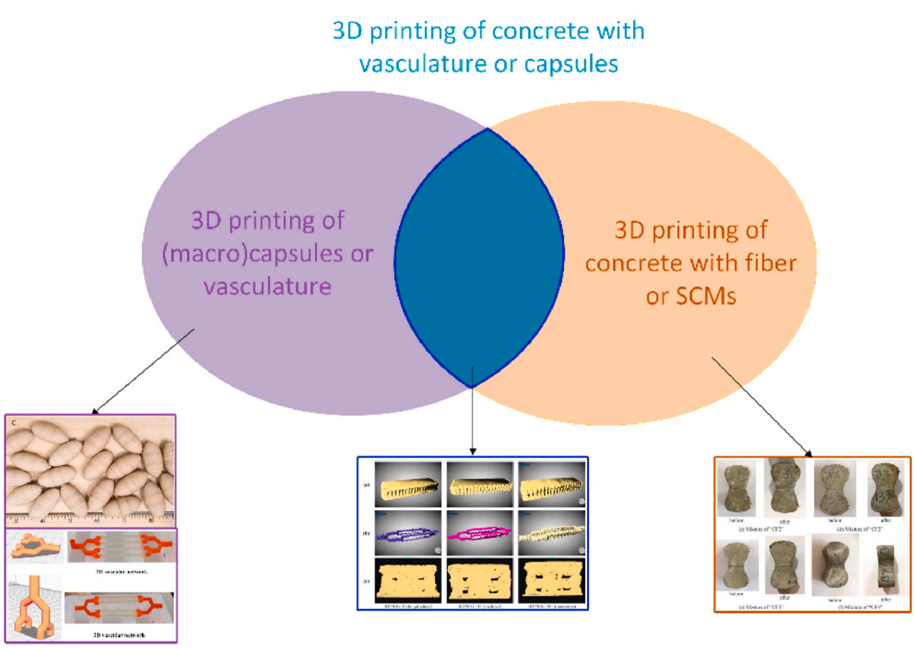


Fig. 4. Strategies of using AM in self-healing cementitious materials.

amount of healing agents in a single capsule. Instead of microcapsules, some researchers used macro-capsules or tabular capsules for better self-healing efficiency. Formia et al., 2015, 2016 first fabricated extruded cementitious tubes with liquid sodium silicate as the healing agent. In their work, an extruder was employed for creating two types of cementitious hollow tubes, i.e., bucatino shape and maccherone shape (Fig. 5a). To increase the watertightness property of the cementitious tubes, they were coated (with a two-component epoxy primer) and sealed (with an epoxy-based two-component thixotropic plaster). The results show that a remarkable strength/stiffness recovery is possible for the samples with maccerone shaped tubes. However, it is mentioned that the production of these hollow tubes is labour intensive. Anglani and his colleagues carried out some further studies on the extruded cementitious capsules (Anglani et al., 2020a). Different kinds of healing agents (a water-repellent agent, a polyurethane precursor, and a solution with bacteria) were used to improve the crack sealing capacity. Furthermore, the performance of the samples with cementitious hollow channels was also investigated when the materials are under static and cyclic loading (Anglani et al., 2020b, 2023).

More recently, the same group focused on fabricating macro-capsules using 3D printing. Novel rigid acrylate macro-capsules and alumina capsules (Fig. 5b) were additively manufactured with a stereolithographic 3D printer, and these two 3D-printed capsules were compared with the extruded cementitious counterpart (Riordan et al., 2023). In the research, three printers were employed to fabricate the macro-capsules. Except for characterizing the 3D-printed shells, the triggering behaviour and self-sealing response of the 3D-printed macro-capsules were evaluated and compared. The results indicate that the 3D-printed macro-capsules are promising due to the favourable healing performance. In a different study, the influence of shell materials was investigated (Anglani, 2019). There, four commercial filament materials, i.e., Poly(lactic) acid (PLA), Polyethylene terephthalate (PET), Polyethylene terephthalate glycol modified (PETG) and Poly (methyl methacrylate) (PMMA) were used to fabricate the capsules (Fig. 5c). The healing performance in terms of strength recovery was investigated when sodium silicate and PU resin were employed as the healing agents. The results show that the capsule system has great potential for prolonging the service life of concrete structures. To fully realize the potential of additive manufacturing, the tailoring of the capsule shape is recommended. Similarly, capsules were fabricated with PLA printing filament and the fracture strengths were numerically simulated (Choi et al., 2021). It was shown that the self-healing materials embedded with 3D-printed capsules have slightly lower compressive strength compared with the one without capsules. This is inferred to be caused by the trapped air bubble between the capsule and the cement matrix.

Except for commercial printing filaments, other materials have been used to fabricate the capsules for self-healing concrete. To balance the trade-off between the survivability of the capsules in concrete mix and the efficient triggering of healing process (rupture of capsule), Sinha et al. (Sinha et al., 2021, 2022a) used a biodegradable material to fabricate the capsules (Fig. 5d). In their work, capsules with different dimensions, aspect ratios (1–2) and shell thickness (0.4–2.0 mm) were investigated in terms of their survivability. Based on the obtained results, it was found that the capsule with an aspect ratio of 1.5, shell thickness of 0.4 mm has the best survival ratio. When using a reagent-grade sodium silicate solution as the healing agent, the cracking strength and bulk resistance are well recovered after 14 days of healing. Furthermore, a novel property-switchable capsule system has also been fabricated using another biomass component, polylactic acid (PLA). It was found that the tensile strength of PLA decreases by over 20% after immersing in alkaline solutions with a pH of 13. However, the degraded PLA shell enables easy rupture for triggering the healing process.

It is worth noting that the aforementioned macro-capsules are only

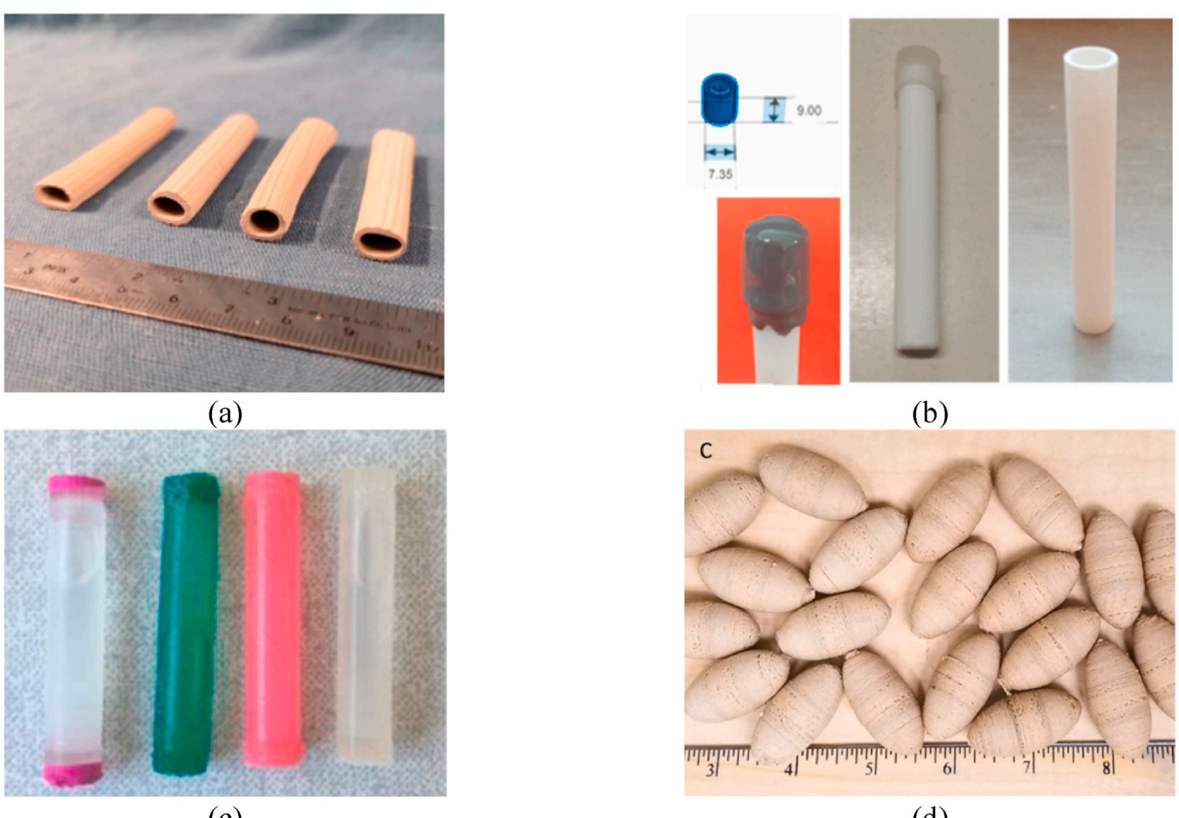


Fig. 5. (a) Extruded cementitious hollow tubes (Anglani et al., 2020b); (b) 3D-printed acrylate/ceramic capsules (Riordan et al., 2023); (c) 3D printed capsules (from left to right PMMA, PET, PLA, PETG) (Anglani, 2019); (d) Lignin-based capsules (Sinha et al., 2022b).

suitable for one-component healing agents. To address this problem, Lim et al. (2023) developed a novel capsule structure, which could be used to host two-component healing agents. As shown in Fig. 6a, the components of the healing agents were injected to the capsules during the fabrication process. Four types of membrane structure were printed using PLA filaments with FDM 3D printing (Fig. 6b). The capsule breaking load and relative standard deviation of the capsules was experimentally and numerically investigated. Based on the results, it is feasible to achieve isotropic strength in capsule manufacturing by modifying the tilting angle and cross-sectional shape of the separator.

2.2. 3D printing of vascular system

Although large volume of healing agents could be stored in the 3D-printed macro-capsule, the discontinuous capsules are still not capable of healing the re-opened cracks. In contrast, the healing agent in a vascular system of cementitious matrix could be replenished, and the vasculature could be used to transport the healing agent to the cracked region for multiple times. Furthermore, a vascular system with complicated geometry could have a higher healing capacity due to the large coverage of healing agent (Shields et al., 2021b). Early studies focused on fabricating customized accessories (such as network connector and reservoir) of the vascular system rather than vascular network itself. For example, Davies et al., 2015, 2021 adopted 3D printing to tailor 3D PLA connector for 2D network configuration (Fig. 7). In their work, a 2D orthogonal vascular network was created with polyolefin tubing, which was later removed due to its high shrinkage ratio under high temperature. To fix the vascular network during concrete casting, connectors were additive manufactured with PLA filament. As shown in Fig. 7d, the connection helps create a dedicated flow channel in the 'joint' of two perpendicular tubes. Furthermore, a 2D bespoke 3D PLA connection is fabricated for wall panels, where a void is designed for placing reinforcement. When using cyanoacrylate as the healing agent, a remarkable strength recovery is observed in a relatively short time scale with the pressurised vascular networks. Moreover, it is demonstrated that the proposed method is scalable and could be applied in the real-scale concrete structures.

To enable supplying healing agents from the outside, Minnebo et al., 2017a, 2017b 3D printed distribution pieces with acrylonitrile butadiene styrene (ABS) and then integrated the pieces into a vascular system (Fig. 8a). As shown in Fig. 8a, there are four tubes connected to the distribution pieces. When cracks occur, the healing agents could be injected from the inlet, which is in the upper part of the printed piece. In their study, 3-point bending and 4-point bending test were carried out to test the healing performance of the vascular healing network. Compared with the reference and the samples without tubes, the vascular ones can undergo more loading cycles. A similar work was carried out by Tsangouri et al., 2019, 2022. There, instead of providing healing agents from the outside, a reservoir is fabricated using PET to house the healing agent for the vascular system (Fig. 8b). The healing performance of samples with different tubes and supply system was investigated. Based on the result from Tsangouri et al. (2022), it is found that the 3D printed polymer networks work as efficient as the ceramic ones. However, the samples with inlet/outlet tubes systems showed better crack sealing capacity and mechanical recovery than the reservoir ones.

More recently, researchers investigated the possibility of fabricating the whole vascular network instead of parts of the system (Vangansbeke et al., 2023). To minimise the turbulence flow at joint, Li et al. (2020) designed the 3D vascular networks based on Murray's law (Murray, 1926). In the work, the designed vasculatures were printed with PLA filament using a commercial 3D printer (Ultimaker, Utrecht, the Netherlands). Except for the 3D vascular systems, 1D- and 2D-structures were also printed and compared with the 3D ones (Fig. 9). From the results, it is noted that the sample with 3D vascular system has better mechanical property recovery (34%) than that of the 1D/2D counterparts (~20%), which is inferred to be caused by the larger coverage of healing agent distribution. Besides, the water absorption of the samples with 3D vascular networks is reported to be reduced by 77%. A vascular network based on Murray's law is fabricated and embedded in a beam of real-life size (0.15 m × 0.24 m x 1.27 m (W x H x L)) (di Summa et al., 2023). However, it is worth noting that PLA may suffer from fast degradation under high pH environment such as cementitious matrix (Xu et al., 2011). Besides, the lack of sufficient bonding between PLA vascular networks and cementitious matrix may introduce a pathway for water ingress (Shields et al., 2022).

Alternatively, other printing materials are employed to create the vascular system for self-healing. Among others, Wan et al. (2022a) created the vascular networks with ABS filaments. There, an octet-shape vasculature was designed and additively manufactured to avoid the possible blockage caused by casting or previous healing events (Fig. 10). The results show that the initial mechanical properties of the samples with 3D-printed vascular system are greatly influenced by the printing parameters such as printing layer-height and printing directions. With a two-component epoxy resin as the healing agent, a full mechanical recovery was observed for all the vascular samples. However, the influence of printing parameters is reduced as the increase of crack-healing cycles. Except for ABS and PLA, another widely used filament is Polyvinyl alcohol (PVA). Due to the water-soluble property, PVA is often used to create the support structure during the printing process. To eliminate the interfacial problems caused by the existence of vascular wall, PVA is used to fabricate the hollow channels, which serves as the pathway for healing agent transportation. Among others, Li et al. (2019) carried out a comprehensive investigation on the feasibility of embedding 3D-printed PVA vasculature for self-healing in the cementitious matrix. They investigated the factors influencing the dissolution of extruded PVA in the highly alkaline environment of the cement paste

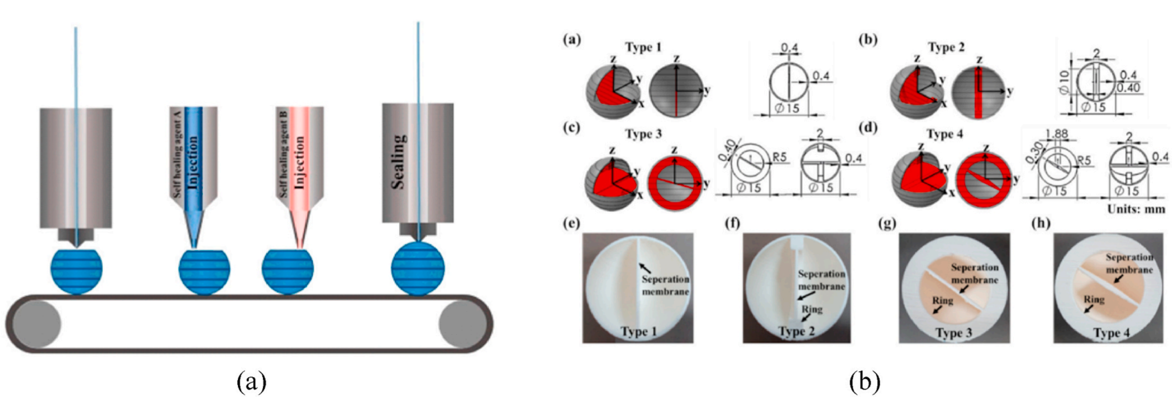


Fig. 6. Capsules that house two-component healing agent (a) fabrication of two-component capsules; (b) design of four types of capsules that house two-component healing agent (Lim et al., 2023).

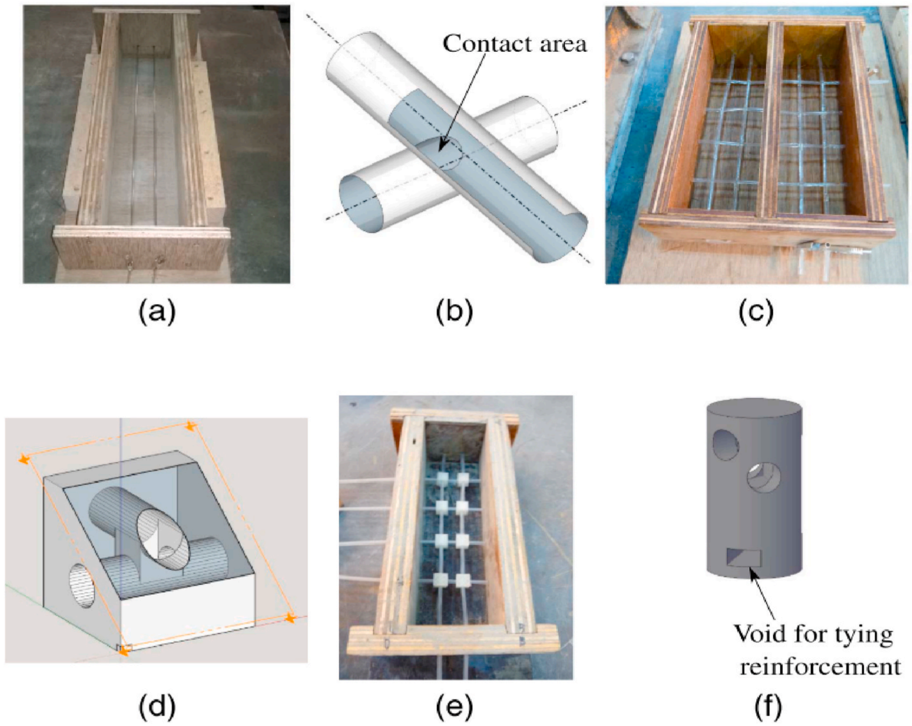


Fig. 7. 2D vascular system in the mould for casting (Davies et al., 2021).

from the chemical aspect. Based on the obtained results, it is found that the main functional groups of the extruded and unextruded PVA is identical. The dissolution of PVA is not influenced by pH and is accelerated by the elevated temperature.

Based on the research of Li et al., Wan et al. (2023a) printed PVA vascular systems for self-healing cement-based materials. There, the dissolution of PVA tubes under different printing configurations was first investigated. To mitigate the volume expansion of PVA during the dissolution process, hollow PVA tubes were printed with different thickness/radius ratio (0.1–0.3). Moreover, the tubes were coated with wax to avoid generating insoluble products from the reaction between PVA and cementitious matrix. The results show that the PVA tubes printed in vertical direction dissolves well when wax coating is applied. After determining the printing configurations, two types of vascular systems were additively manufactured (Fig. 11). Although the vascular systems adversely affect the initial flexural strength, the mechanical recovery after each healing process is over 100% compared with the original specimens. Compared with 2D vascular system, 3D vascular system allows more crack-healing cycles.

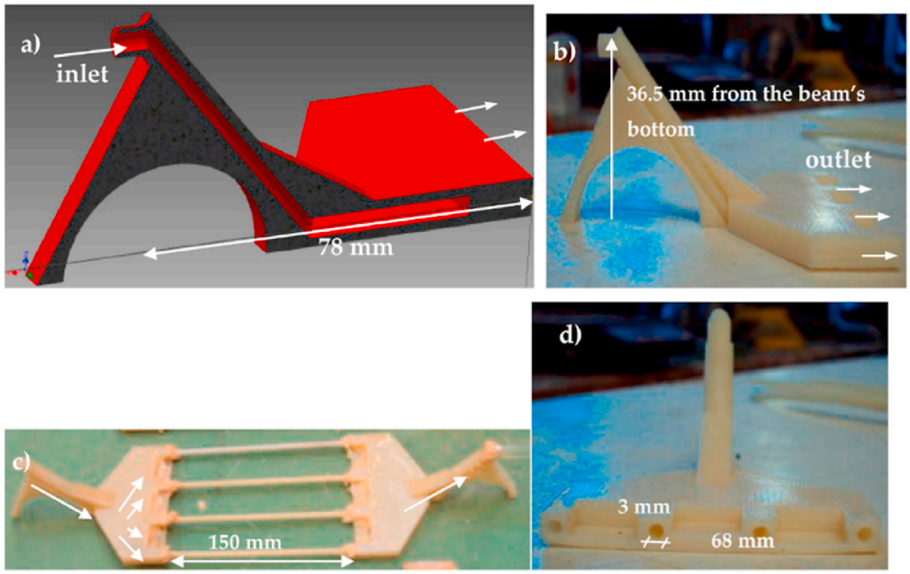
As mentioned above, the existence of vascular system reduces the initial mechanical properties of the cement-based materials. De Nardi et al. (De Nardi et al., 2020, 2021) additive manufactured tetrahedral mini-vascular networks (MVNs) with PLA filament for self-healing concretes. Except for the general requirements for vasculatures in self-healing concrete (i.e., survival of the mixing process and timely rupture when crack occurs), the influence of vasculatures on the cementitious matrix is considered when designing the MVNs. In particular, the tetrahedral shape is chosen considering its proneness to crack when hit by cracks compared with the spherical shape. The size of MVNs is chosen to be twice the size of the coarse aggregates. A schematic of the designed MVN unit is shown in Fig. 12a. In addition, the bond between the MVN ligaments and host materials is further strengthened by including ribs. It is observed that the average initial flexural strength of samples with MVNs decreases by 9–16%. However, the strength and stiffness are regained by 20% and 75–80% respectively when using liquid sodium silicate in the MVN as the healing agent. Moreover, the same research group further developed the MVNs to store two-component healing agents (De Nardi et al., 2023). Specifically, tetrahedral shaped units (TETs) with coaxial hollow ligaments (d-TETs) are additively manufactured with PLA printing filaments (Fig. 12b). According to the study, the healing process was successfully triggered when the crack-width reached 0.35 mm, and remarkable recoveries in terms of strength and stiffness are achieved with several types of healing agents (Lim et al., 2022).

2.3. Characterization of 3D-printed capsules/vasculature

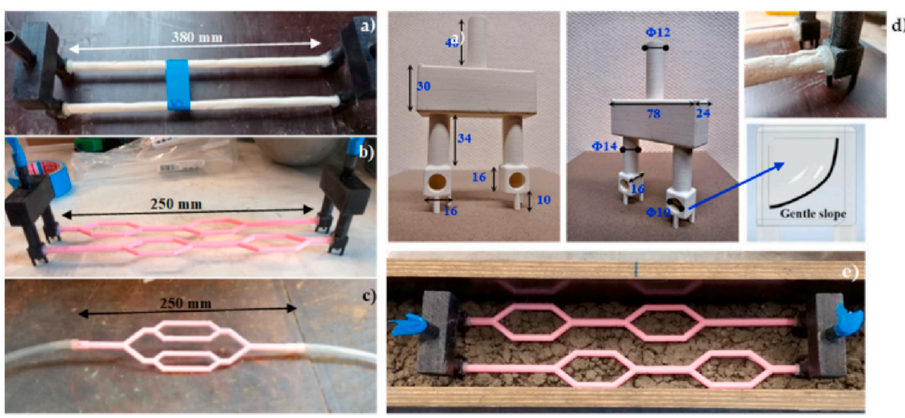
The 3D-printed capsules or vasculature needs to be timely ruptured for triggering the self-healing process. Therefore, it is of great importance to characterize the 3D-printed capsules or vascular networks. According to the previous studies, there are mainly two characterization aspects.

- Shell continuity/water tightness
- Fracture behaviour

For 3D-printed vascular network, good water tightness of vascular networks avoids the blockage of vascular system during concrete casting (Wan et al., 2022a). While for 3D-printed macro-capsules, the capsules need to have good shell continuity to house the healing agents before the occurrence of crack. For the capsules with poor watertightness induced by 3D printing, it is necessary to include additional protective layers to prolong the shelf time of the healing agents (Riordan et al., 2023). Except for increasing the shell continuity, the coating layers could also help increase the bonding strength between capsules and cementitious matrix (Anglani, 2019). Except for the intrinsic "defects" from the 3D printing process, the harsh environment (i.e., high alkaline) in the host materials may cause degradation of capsules (especially printed with PLA). Choi et al. (Choi et al., 2021) increased the stability of 3D-printed PLA capsules by coating lacquer and resin. Solubility tests were performed on three groups submersed in a $Ca(OH)_2$ solution with a pH of 13. A similar study was carried out by Sinha et al. (2021), where the alkaline degradation of capsule shell materials was investigated. The results show that the degradation of PLA capsule in the first 28 days in



(a)



(b)

Fig. 8. (a) 3D printed distribution pieces (Minnebo et al., 2017b); (b) Reservoir-vascular networks (Tsangouri et al., 2022).

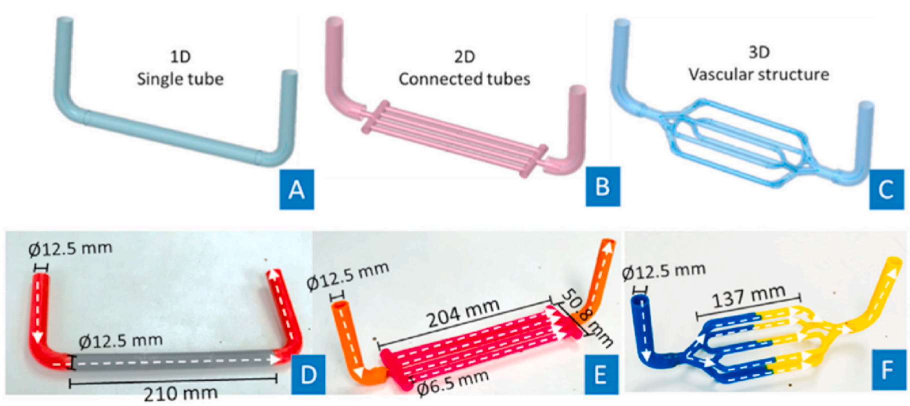


Fig. 9. Schematics of designed 1D/2D/3D vascular structures (Li et al., 2020).

negligible, while serious degradation was observed after 90 days.

The fracture behaviour of 3D-printed capsules or vasculature is also of great importance. Some researchers tested the mechanical properties of the printed items prior to embedding them to the host materials. On one hand, the mechanical experiments towards capsules help optimize the configurations (such as wall thickness) of capsules for balancing the survivability and rupture behaviour (Anglani, 2019). For example, Li et al. (2020) compared the load response of 1D/2D/3D printed vascular

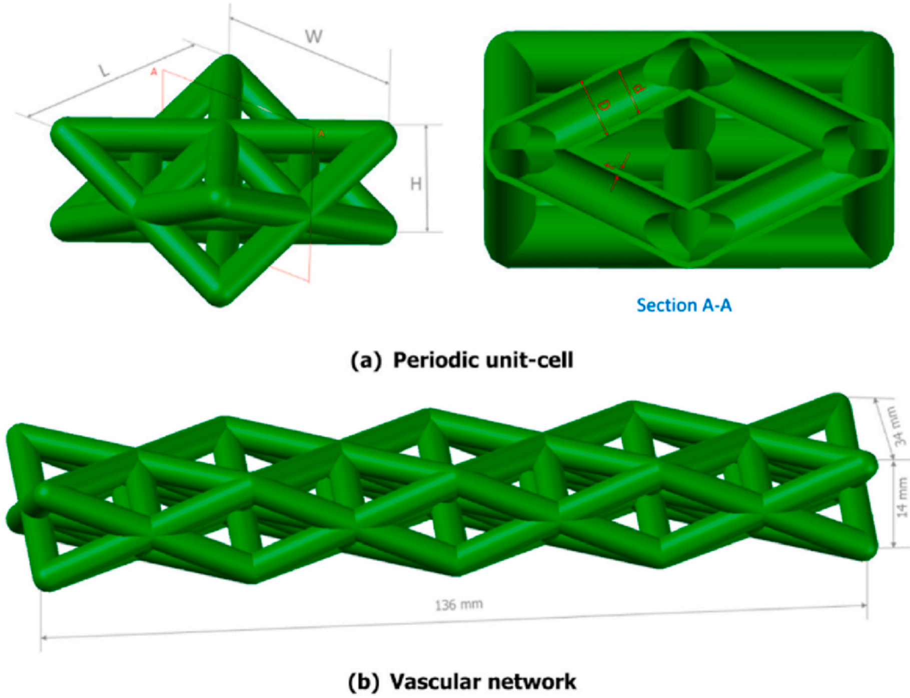


Fig. 10. Schematics of 3D-printed ABS vascular networks (Wan et al., 2022a).

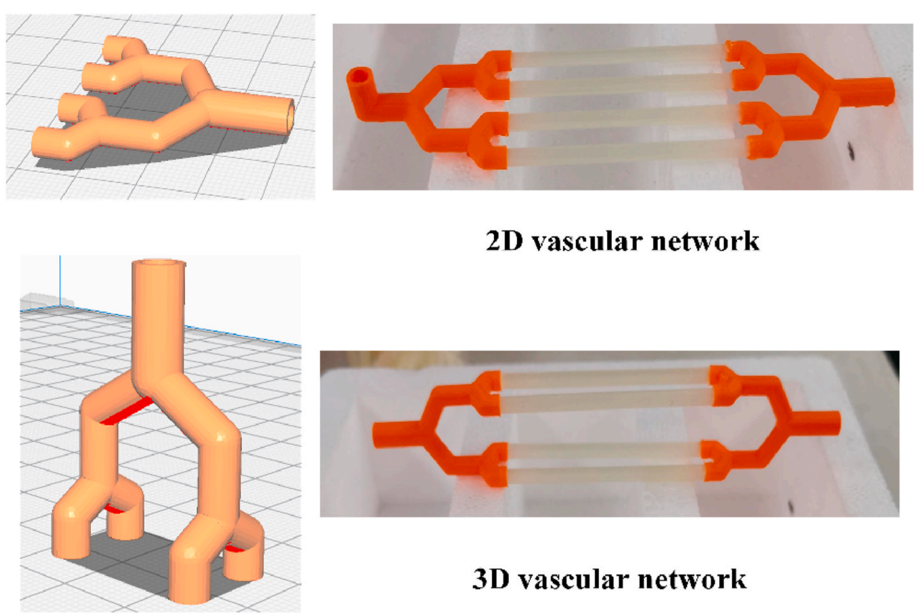


Fig. 11. Schematics of 3D-printed PVA vascular networks (Wan et al., 2023a).

structures and found that all the printed structures fail at a low load, making the 3D-printed vasculatures suitable for self-healing concrete (Fig. 13). On the other hand, the input parameters for numerical simulations could be accessed by the characterization of the capsules/vasculature, which helps understand the self-healing cementitious materials with 3D-printed capsules/vasculatures (Anglani et al., 2020b; Zhou et al., 2022).

3. Self-healing in 3DCP

Except for printing additional accessories (capsule or vasculature) using thermoplastic materials, 3D printing of concrete is another option. Similar to the mould cast self-healing concrete, there are mainly two approaches to achieve self-healing in 3D printed concrete, i.e., through autogenous healing and autonomous healing.

3.1. Autogenous healing in 3DCP

To achieve a good ductility of 3D printed concrete, an increasing number of studies focuses on 3D printable engineered cementitious composites (3DP-ECC) (van Overmeir et al., 2023; Zhou et al., 2022; Yu et al., 2021). As mentioned above, adding fibres in cementitious materials promotes the self-healing by inhibiting crack propagation and thereby promoting the autogenous healing (Nguyen et al., 2023).

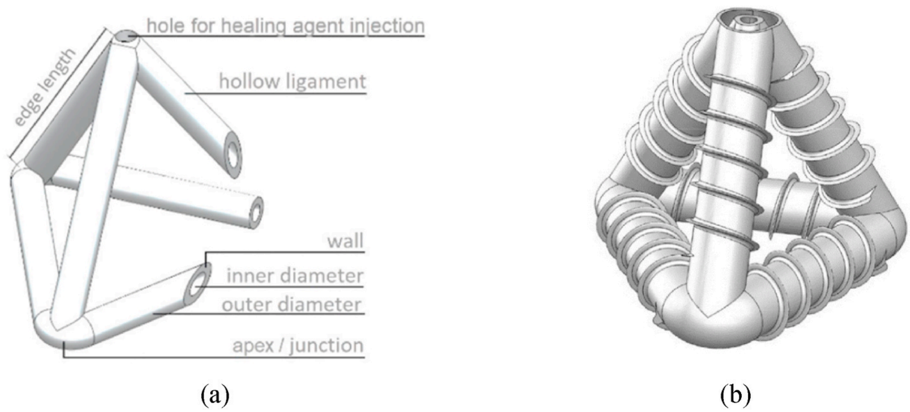


Fig. 12. (a) Schematics of a MVN unit (De Nardi et al., 2020); (b) d-TET housing two-component healing agents (De Nardi et al., 2023).

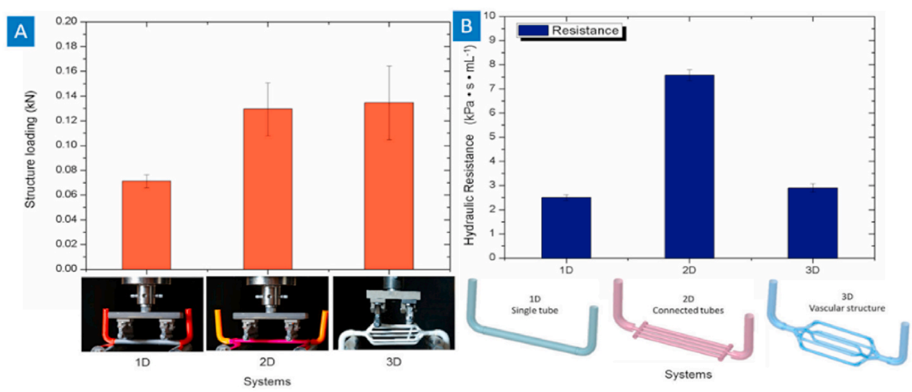


Fig. 13. Reconstructed CT results of 3D-printed vascular self-healing cementitious materials (Li et al., 2020).

Fig. 14. Crack sealing of 3DCP with steel fibre coated with PVA and GO (graphene oxide) (Ahmadi et al., 2023).

However, a large percentage of fibre may negatively influence the properties of the 3DP-ECC due to the air void (van Overmeir et al., 2022). Ahmadi et al. (Ahmadi et al., 2023) focused on reducing the amount of fibres without influencing the mechanical and self-healing properties of 3D printable concrete. In their work, the micro steel fibres were coated with the solution of PVA and graphene oxide (GO). Compared with 3D printed steel fibre reinforced concrete, the self-healing performance of the recommended material is further enhanced. As shown in Fig. 14, crack closure was observed for the designed 3D printable materials after a long healing period (105 days of wet-dry cycle). Besides, the maximum tensile and compressive strength recoveries were reported as 34.4% and 43.9% measured by direct tensile test and concrete compressive test, respectively.

Other studies focused on the influence of curing conditions on the self-healing efficiency of 3DP-ECC. Among others, Fernández et al. (2023); Jarabo et al. (2023) investigated the self-healing behaviour of 3DP-ECC in two curing environments, i.e., room temperature (34 ± 2% RH, 20 ± 2 °C) and curing chamber (98 ± 2%RH, 20 ± 2 °C). In their work, PVA fibres of 8 mm length were incorporated to the mixture to form ECC with a volume percentage of 1.5%. The results show that the material in curing chamber can seal the cracks induced during the loading tests after 28 days (Fig. 15). Ma et al. (Ma et al., 2022) focused on the durability performance of 3D-printed concrete in the sulfate-attack environment. The printing ink is a sulfoaluminate cement-based composite with polypropylene (PP) fibres. From the experimental results, it is observed that the self-healing properties of sulfoaluminate cementitious composites helps reduce the damage anisotropy of 3DCP.

Except for concrete, 3D printing of geopolymer is also widely investigated to address the environmental issues caused by cement production (Raza et al., 2022). Some researchers investigated the self-healing capacity of 3D printed geopolymer. For example, Tang and Tang (2022) additively manufactured the gradient diamond/-geopolymer composites (DGP) using an extrusion-based equipment. In their study, two types of ink were prepared with potassium silicate solution, NaOH, KH_2PO_4, diamond and geopolymer powder. Due to the addition of diamond, part of the pores was healed during the reaction (Fig. 16). From the results, it is found that the geopolymers with 15 wt% diamond could effectively avoid cracks in both early age and high temperature conditions. Therefore, it is feasible to adopt diamond as rheology modifier and promote the self-healing of DGP.

3.2. Autonomous healing in 3DCP

For self-healing concrete embedded with a 3D-printed vascular system, one problem is that the orientation of the vasculature may change during concrete casting compared to the designed places due to the vibration or buoyancy force (Wan et al., 2023a). Alternatively, in-situ fabrication of vascular system with additive manufacturing may help alleviate the problems. Zhang et al. (Zhang et al., 2022) developed a programmable construction of vascular system for self-healing by 3D printing technology. The influence of ink composition, printing parameters, and printing time was investigated. In particular, the cementitious material matrix serves as the vessel wall for the printed vasculatures, allowing for programming into different geometries. The fugitive ink was prepared with sodium alginate (ALG) emulsions. In-situ vasculature construction was performed with the moulded fresh cement paste as supporting matrix. The cement paste with printed vasculature was demoulded and cured in saturated $Ca(OH)_2$ solution for 28 days. A hollow vascular system was formed after the release of methyl palmitate (MPA) and contraction of printing filaments. The schematic could be found in Fig. 17. When the healing agent (liquid sodium silicate) was pumped into the 3D-printed for 10 days, the water absorption of the healed samples was reduced by about 70% compared with the references, indicating the remarkable healing performance of the proposed construction of vascular self-healing cementitious materials.

In addition, considering the fabrication (and the possible post-processing) of vascular system is time-consuming and labour-intensive, direct ink writing of vascular self-healing cementitious materials was investigated by some researchers. Among others, Wan et al. (2023b) additively manufactured the vascular self-healing cementitious materials by intentionally reserving hollow channels in the host materials. They first developed a 3D printable cementitious ink incorporated with 0.17 vol% PE fibres. After testing the rheological properties of the designed mixture, the vascular self-healing cementitious materials were fabricated with the top and bottom layers printed in different printing direction. The results from CT scanning of the designed vascular self-healing samples are shown in Fig. 18. The macrostructure and healing performance in terms of mechanical and watertightness were compared. The results indicated that more than 40% of the hollow channels was compressed by the upper layers during the printing process. Although a remarkable recovery of mechanical property and watertightness was observed in the test, printing direction significantly influences the properties of the 3D-printed vascular self-healing cementitious materials.

4. Future challenges of AM in self-healing cementitious materials

Although great progress has been made for the application of additive manufacturing in self-healing cementitious materials (Šavija, 2020, 2023), there are still several aspects to be further addressed in the future.

The vascular networks in self-healing concretes have not been subject to optimization in terms of shape to optimize the overall performance. One of the advantages of additive manufacturing is the possibility of creating arbitrary shapes of vascular networks. Although the shape of 3D-printed vascular system is more complicated than the traditional ones, they are still far simpler compared to e.g., leaf venation. Furthermore, the geometry of vascular networks could be tailored for different structural members. For example, more vasculatures could be arranged in the places that have higher stress or are susceptible to

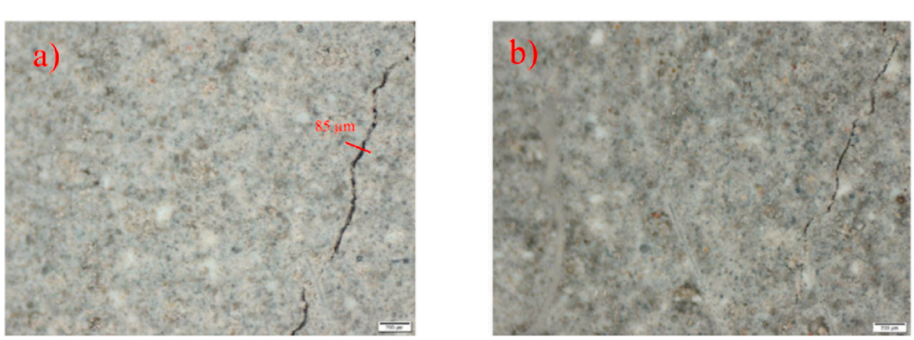


Fig. 15. Preloaded crack in the 3D-printed ECC in curing chamber. (a) 2 days; (b) 28 days (Fernández et al., 2023).

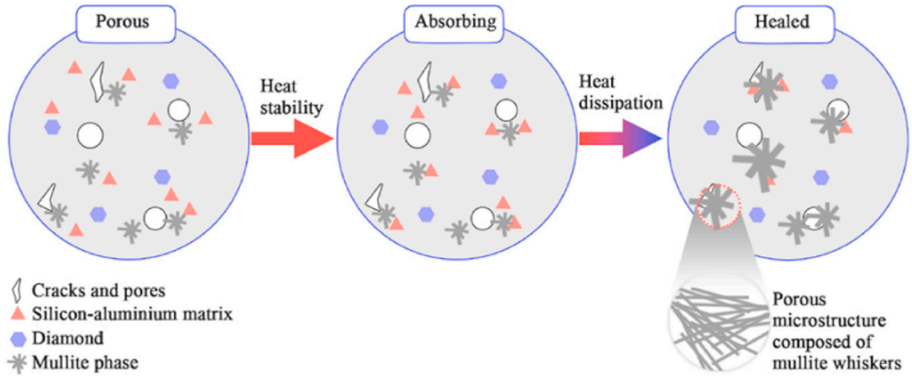


Fig. 16. Schematic of self-healing mechanism in DPG (Tang and Tang, 2022).

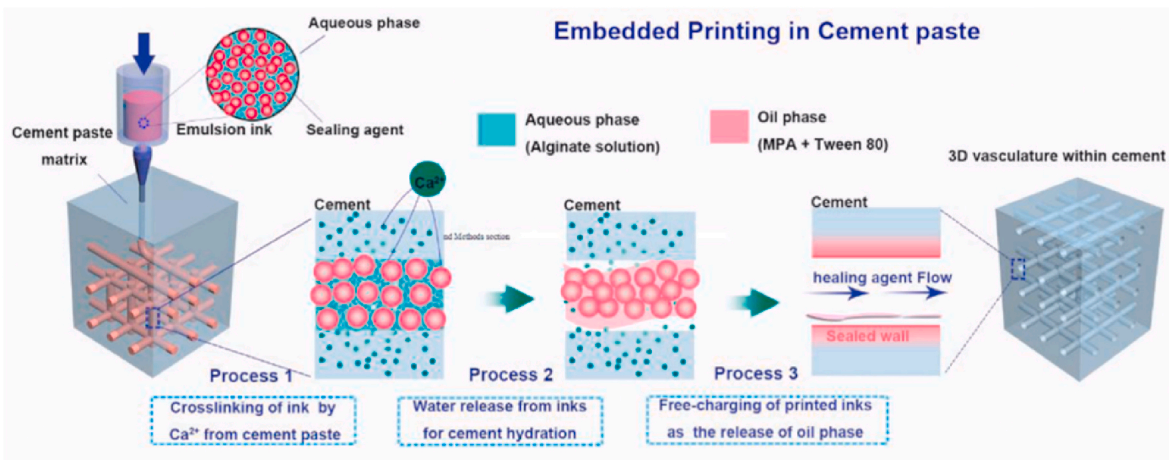


Fig. 17. Schematic of 3D printing of vasculature in cementitious matrix (Zhang et al., 2022).

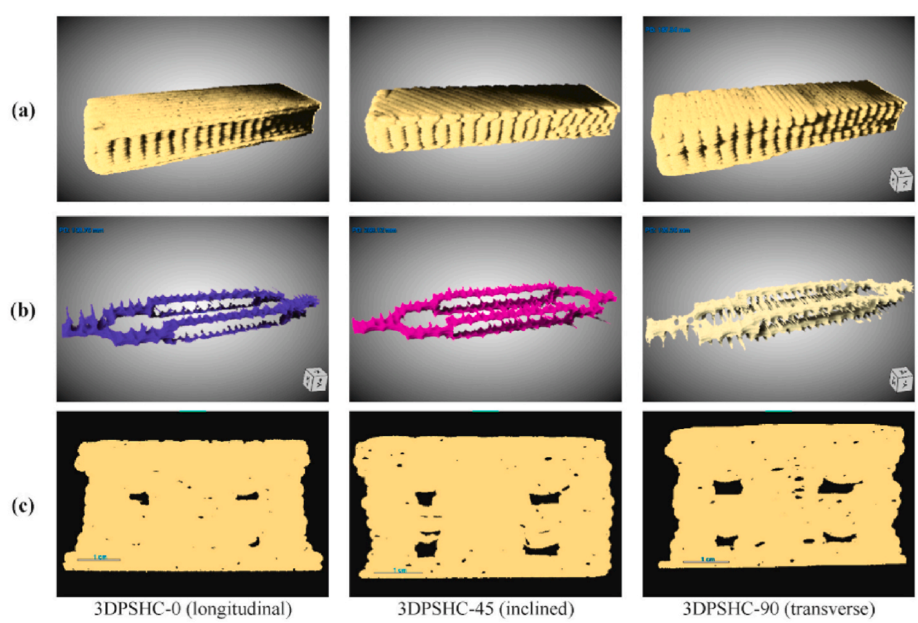


Fig. 18. Reconstructed CT results of 3D-printed vascular self-healing cementitious materials (Wan et al., 2023b).

cracking in the structural members. Besides, the reinforcement system could be taken into consideration when designing and fabricating the vascular system. Except for trial-and-error approaches, methods such as evolutionary algorithms and machine learning (ML) have been employed to optimize the structures under certain configurations (Wan et al., 2022b; Sui et al., 2021; Chen and Gu, 2020). Since ML is data-hungry, it is necessary to develop reliable numerical models for generating the dataset for optimization task (Gupta et al., 2021; Xi et al., 2023). Although some researchers numerically investigated the self-healing concrete with vasculatures (Gardner et al., 2014; Sayadi et al., 2023) or capsules (Lv et al., 2017), the mapping relationship and computational time need to be considered when using these models for dataset generation (Wan et al., 2023c).

The scalability of self-healing cementitious materials with 3D-printed capsules or vascular networks must also be addressed in the future research. Due to the limitations of commercial printers, the dimension of 3D-printed vasculatures in published literature is less than 500 mm, making it difficult to apply in real-life structures. Although it is possible to print the vascular network in pieces and then ensemble into vascular system (Tsangouri et al., 2019; di Summa et al., 2023; Davies et al., 2018; Teall et al., 2016), this is time-consuming and labor intensive. The direct ink writing of vascularized self-healing concrete has great potential for upscaling. For example, modularized and standardized 3D-printed concrete elements with vascular system could also be applied in real-scale concrete structures. But further investigation is needed for the design of vascular system and the mix design of cementitious ink. It is worth noting that two-component strategy could be a good solution to narrow the shape difference between designed and printed channels.

The life cycle assessment (LCA) and life cycle costs (LCC) should be further studied in the future. Based on previous research (Garces et al., 2021), it is found that self-healing geopolymer concrete may be better in terms of global warning potential compared with ordinary concrete. The usage of additive manufacturing in self-healing concrete could help to realize the full potential of self-healing through the fabrication of more complicated capsules or vasculatures. Compared with microcapsules, the quality of 3D-printed capsules/vasculatures is more controllable. However, there is currently limited research on the additional costs induced by the digital fabrication of capsules or vasculatures. Furthermore, the long-term durability of self-healing concrete with 3D-printed capsules/vasculatures in the real environment has not been investigated (di Summa et al., 2023; Rengaraju and Al-Tabbaa, 2023; Kannikachalam et al., 2023). Therefore, it is necessary to further investigate the LCA and LCC.

5. Summary and conclusions

This review summarized and discussed published literature related to the application of 3D printing in self-healing cementitious materials. In accordance with the possible functional strategies for self-healing in cement-based materials, there are three main strategies for incorporation additive manufacturing (AM) in self-healing cementitious materials, i.e., self-healing concrete with 3D-printed capsules or vasculatures, 3D printing concrete with fibers or supplementary cementitious materials (SCMs) and their combination. The first approach, especially 3D printing of vascular system, has been widely investigated in the previous studies. Compared with conventional vascular/capsule based self-healing cementitious materials, more healing agent could be housed, enabling the healing large of wider cracks. However, due to printer limitations, the fabrication of capsules or vasculatures is time-consuming. 3D printing of concrete could be an option. Considering the current absence of reinforcement or coarse aggregates in 3DCP, fibers are often included to increase the ductility of 3DCP. Besides, (novel) SCMs are involved in the printable cementitious ink, which may also promote the autogenous healing of cementitious materials. Last, encapsulation or vascularization of healing agents could be included to further enhance the autonomous healing of 3DCP.

However, there are still several challenges to be addressed in the future. To fully realize the potential of 3D printing, the shape of capsules and vasculatures should be optimized. One of the promising solutions is data-driven optimization through the combination of machine learning (ML) and numerically generated dataset.

Another issue is the scalability of the self-healing cementitious materials fabricated using additive manufacturing. Among the three approaches, the last one (3DCP with vasculature or microcapsules) shows the great potential for the real-life concrete structures. Furthermore, life cycle assessment (LCA) and life cycle costs (LCC) could also be further investigated.

CRediT authorship contribution statement

Zhi Wan: Conceptualization, Data curation, Investigation, Visualization, Writing – original draft. **Yading Xu:** Data curation, Writing – review & editing. **Shan He:** Data curation, Writing – review & editing. **Erik Schlangen:** Project administration, Supervision, Writing – review & editing. **Branko Šavija:** Project administration, Supervision, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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