A REVIEW ON SELF-HEALING IN REINFORCED CONCRETE STRUCTURES IN VIEW OF SERVING CONDITIONS

Haoliang HUANG[1, a] and Guang YE[1,2,b]

[1]Department of Civil Engineering and Geoscience, Delft University of Technology, the Netherlands

[2]Department of Structural Engineering, Ghent University, Belgium

[a]haoliang.huang@tudelft.nl, [b]g.ye@tudelft.nl

Abstract: In this paper, different mechanisms of self-healing, i.e. self-healing based on adhesive agents, self-healing based on bacteria, self-healing based on autogenous self-healing were described. Their required conditions were summarized. The previous investigations showed that all mechanisms of self-healing are effective to some extend under particular conditions. In this paper, concrete structures were categorized according to serving conditions. Potential self-healing mechanisms are pointed out according to the required conditions of each self-healing mechanism.

Keywords: Self-healing, concrete structures, serving conditions, design

1. INTRODUCTION

Damage Prevention Paradigm is a traditional design philosophy that focuses on preventing damage by using stronger materials [1]. Professor J.E. Gordon [2] gave a clear picture on this design principle in his book “Structures: or why things do not fall down”. This principle is mostly used in current engineering material design [1]. Regarding the design of reinforced concrete structures, designers determine the amount of steel reinforcement added in each element based on the principle of damage prevention, explaining the question “why do not you fall through the floor”. However, concrete is always designed to crack to make the steel reinforcement adequately carry tensile loads. Moreover, microcracks can even occur throughout the material body prior to application of any load [3]. These cracks are not considered as damage as long as a prevailing crack width criterion is not exceeded [3]. Therefore, a design of reinforced concrete is based on Damage Control Paradigm [3]. Although these cracks are not considered as damages in reinforced concrete structures, they are not desirable in view of the resistance against the ingress of harmful substances. It is well known that cracks provide preferential accesses for aggressive agents, such as chloride, sulphate and carbonate. These aggressive agents can not only induce corrosion of reinforcement steel, but also degrade the concrete. Both the corrosion of reinforcement bars and the degradation of concrete can reduce the service life of reinforced concrete structures.

For this service life problem caused by cracks, man-made repair is a common solution [4]. Although man-made repair can prolong the service life of reinforced concrete structures, it has some important drawbacks. For instance, the cost of man-made repairs are usually very high [5]. It was reported that the total amount of money involved in repairing and upgrading

America's bridges is 140 billion US dollars [3]. In United Kingdom, almost 45% of the budget for UK's activity in construction and building industry was spent on repair and maintenance [3]. Moreover, if structures, like bridges and tunnels, have to be taken out of service for repair, the indirect costs are generally ten times higher than the direct costs [4]. Apart from the high cost of repair, it is recognized that realizing durable repairs is difficult, even though the quality of such repairs has substantially increased in recent years. Most of these repairs can only last for ten to fifteen years [4]. Furthermore, it is difficult to repair cracks which are not accessible, such as the cracks in underground concrete structures [6].

In comparison, self-healing of cracks in concrete structures could be beneficial [6]. As a novel idea, self-healing of cracks has attracted much attention in recent years. Reinforced concrete structures should be made smart enough to detect their own damage and repair themselves [7]. These self-healing concrete structures have significant potential to extend their service life and reduce their economic, social and environmental costs [7].

The potential application (design) of self-healing in reinforced concrete structures is discussed in this paper. The advantages and disadvantages of various mechanisms of self-healing are summarized. These mechanisms of self-healing include autogenous healing, self-healing based on adhesive agents, self-healing based on (expansive) minerals and self-healing based on bacteria. For the reinforced concrete structures exposed to particular environment, i.e., the structures under water, under ground and in the open air, the feasibility of each mechanism of self-healing is analyzed and compared. Possible problems involved in the realization of self-healing in the structures are pointed out and the corresponding solutions are proposed based on the research so far. In this way, this paper provides some reference for realizing self-healing in reinforced concrete structures in future.

2. MECHANISMS OF SELF-HEALING AND REQUIRED CONDITIONS

Based on the literature survey, self-healing of cracks in cementitious materials can be grouped into four categories:

1) self-healing based on adhesive agents,
2) self-healing based on bacteria,
3) self-healing based on mineral admixtures,
4) autogenous self-healing.

The physico-chemical healing process, influencing factors, advantages and disadvantages of these categories of self-healing are described in this section.

2.1 Self-healing based on adhesive agents

The hardening of adhesive agents in cracks has the potential to seal the cracks and connect both crack surfaces. These adhesive agents can be either an one-component or two-component agent, or even a multi-component agent. The hardening processes vary with the properties of the agent. Various adhesive agents applied for self-healing in recent studies are discussed below.

Epoxy is one of the adhesive agents often used for self-healing. In some investigations, a two-component epoxy was encapsulated and pre-embedded in concrete for self-healing [8-10]. When the capsules became intersected by cracks, both components of the epoxy were released and mixed with each other. These two mixed components then reacted and hardened to heal the cracks. However, the reaction can be negatively influenced as the ratio of these two components leaking into the cracks could not be controlled and the optimum mixing could not be achieved [6]. A one-component epoxy has been reported to be more effective for self-healing. For instance, Thao et al. [11] embedded one-component epoxy in concrete by using sealed glass tubes. After the glass tubes were broken by cracks, the one-component epoxy leaked into the cracks. Once this type of epoxy was exposed to air, it started to harden and finally repaired the cracks effectively. Nishiwaki et al. [12] also reported positive results of self-healing by using one-component epoxy.

Methylmethacrylate (MMA) is another type of adhesive agent with two or multi components that can be utilized for self-healing. Yang et al. [13] used microcapsules to supply methylmethacrylate and triethylborane (TEB) to cracks for self-healing. Dry and McMillan [14] even used three-component MMA for self-healing in concrete. In their study, in order to supply this multi-component MMA to cracks, a multi-channel vascular system was built inside concrete. The liquid components were delivered to cracks through this vascular system from the outside (More details can be seen in Section 3.2). Compared with the two-component epoxy, MMA has a lower viscosity and thereby the mixing of components is better. Nevertheless, because the viscosity of MMA is low, it can easily leak out of the cracks. This leaking may decrease the efficiency of self-healing. To overcome this problem, poly methylmethacrylate (PMMA) with higher viscosity was applied to replace MMA as a healing agent [6].

Also, *cyanoacrylate* has been employed as adhesive agent for self-healing. Cyanoacrylate has a low viscosity and is able to penetrate into the matrix. As a result it can connect the crack surfaces strongly [6, 15]. Furthermore, as a one-component adhesive agent, it can harden within a short time after exposure to air [6]. Therefore, cyanoacrylate is frequently used to develop self-healing concrete [16-22]. Apart from the aforementioned agents, other healing agents such as silicon [23] and tung oil [5] were also used.

In general, the efficiency of self-healing based on adhesive agents mainly depends on the type of adhesive agents. One-component agents are easier to operate with and their efficiency is higher than that of two or multi component agents [6]. Usually, adhesive agents make both crack surfaces bond strongly and, therefore, lead to good recovery of mechanical properties of concrete [6].

In addition, one of the important requirements for this self-healing mechanism is the successful supply of adhesive agents to cracks. These liquid adhesive agents are generally supplied to cracks by using capsules or vascular systems. Detailed information about the methods to supply liquid healing agent to cracks will be presented in Section 3.

2.2 Self-healing based on bacteria

The idea of bacteria-based self-healing is to utilize bacteria to promote precipitation of calcium carbonate in cracks. In 1990s, Gollapudi et al. [24] suggested to use bacteria to induce the precipitation of calcium carbonate ($CaCO_3$) to repair cracks. The precipitation of calcium carbonate can be caused by various metabolic pathways, such as the hydrolysis of urea and the oxidation of organic acids [6].

Compared to the other pathways for generating carbonate, the hydrolysis of urea has several advantages. For instance, it can easily be controlled and it has the potential to produce high amounts of carbonate within a short time [25]. Catalyzed by urease, urea is degraded to carbonate and ammonium. The concentration of carbonate increases in the bacterial environment [25]. One mole of urea is hydrolyzed intracellularly to one mole of ammonia and one mole of carbonate (Equation 1), which spontaneously hydrolyzes to one mole of ammonia and carbonic acid (Equation 2) [25]. These products subsequently reach the equilibrium in water to form bicarbonate and two moles of ammonium and hydroxide ions (Equation 3 and 4) [25]:

$$CO(NH_2)_2 + H_2O \Rightarrow H_2COOH + NH_3 \tag{1}$$

$$NH_2COOH + H_2O \Rightarrow NH_3 + H_2CO_3 \tag{2}$$

$$2NH_3 + 2H_2O \Leftrightarrow 2NH_4^+ + 2OH^- \tag{1}$$

$$2OH^- + H_2CO_3 \Leftrightarrow CO_3^{2-} + 2H_2O \tag{2}$$

In the presence of calcium ions, calcium carbonate is deposited, once a certain super-saturation level for the precipitation of calcium carbonate is reached [25]. As shown in Figure 1, because of the negative charge of the cell wall, calcium ions are attracted (Equation 5). As a result, the crystals of calcium carbonate precipitate on the bacterial cell (Equation 6) [6, 25]. In addition, precipitation also takes place in the bulk liquid phase [25]:

$$Ca^{2+} + Cell \Rightarrow Cell\text{-}Ca^{2+} \tag{3}$$

$$Cell\text{-}Ca^{2+} + CO_3^{2-} \Rightarrow CaCO_3 \tag{4}$$

Another metabolic pathway to produce calcium carbonate is the oxidation of organic acids. As expressed in Equations 7, 8 and 9, calcium carbonate is formed with the conversion of calcium acetate during the bacterial metabolism [26]:

$$Ca(CH_3COO)_2 + 4O_2 \Rightarrow Ca^{2+} + 4CO_2 + 2H_2O + 2OH^- \tag{5}$$

$$CO_2 + OH^- \Leftrightarrow HCO_3^- \tag{6}$$

$$2HCO_3^- + Ca^{2+} \Rightarrow CaCO_3 + CO_2 + H_2O \tag{7}$$

Compared to the hydrolysis of urea, which produces excessive ammonium, the oxidation

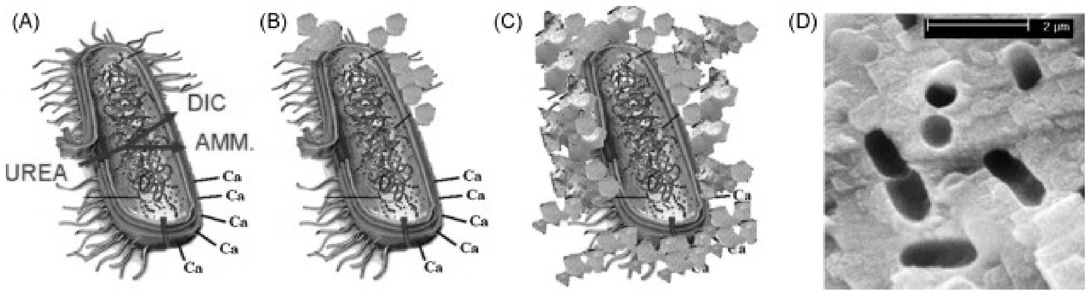

Figure 1: Simplified representation of carbonate precipitation induced by the hydrolysis of urea with the help of bacteria [25]

of organic acids has less environmental impact [27]. Moreover, as presented in Equation 9, during the precipitation of calcium carbonate through this metabolic way, CO_2 is produced as well. The produced CO_2 can also react with portlandite ($Ca(OH)_2$), which is quantitatively an important hydration product of Portland cement, to form more calcium carbonate [28, 29].

However, in order to use bacteria to heal cracks in concrete, some technical problems have to be solved. The bacteria should be protected not only against the alkaline environment in concrete, but also against the decreasing space in the matrix when hydration of cement proceeds [6, 28]. As found by Jonkers [28], when the bacteria spores, which can be viable for up to 50 years, were directly added into concrete, their lifetime dramatically decreased to only a few months. This is caused by the hydration of cement grains. As the cement grains hydrate, most of pores becomes smaller than bacterium spores with the size of 1 μm, which causes the cell to collapse [28]. It is demonstrated that the immobilization of bacteria in porous clay aggregates before the mixing of concrete can prolong the lifetime of bacteria enormously. Self-healing can be triggered later on when cracks intersect these clay particles [28-30]. Contemporarily, researchers in Ghent University are trying to immobilize bacteria in silica gel or polyurethane [31]. The "food" for the bacteria, urea or organic acids, should also be embedded in the matrix in similar ways. It should be mentioned that the essential condition for bacteria-based self-healing is that water is present in cracks. Otherwise the chemical reactions in Equation 1 to 9 can not take place.

2.3 Self-healing based on mineral admixtures

Self-healing can be attributed to reactions of mineral admixtures in cementitious materials. These mineral admixtures are added to the concrete mixture during mixing. After the concrete cracks, some unreacted mineral admixtures are present at crack surfaces. When water penetrates into the cracks, these mineral admixtures start to react with the water in cracks. The cracks are then expected to be filled with reaction products.

By now, the mineral admixtures for self-healing can be categorized into two groups: expansive additive and crystalline additive [32]. In the case of expansive additive, the volume of the reaction products is larger than that of the admixture itself and the expansion depends on the composition of the admixtures [33]. Regarding the crystalline additive, its components can react with $Ca(OH)_2$ to form crystalline products [32]. Both additives have been investigated. For instance, Kishi and co-workers [34, 35] used a mixture of expansive agents, i.e. calcium sulfoaluminate ($Ca_4(AlO_2)_6SO_4$), free lime (CaO) and anhydrite ($CaSO_4$). In their studies, another expansive additive called geo-material, which mainly consists of silicon dioxide, sodium aluminum silicate hydroxide and montmorillonite clay, was also studied [35]. Similar to Kishi's research in [34-36], various types of minerals as expansive admixtures for self-healing were also investigated by Sisomphon [32]. The effects of the mixing ratio of the mineral compositions on the capacity of self-healing were explored as well [32]. Apart from expansive additives, a synthetic cementitious material called "crystalline additive", which contains reactive silica and some crystalline catalysts, was also applied for self-healing [32]. It has been reported that these mineral admixtures lead to remarkable improvement of watertightness of cracks due to self-healing [32, 34-36].

There are some advantages by using mineral admixtures for self-healing. Since some minerals are able to react intensively with water, self-healing of crack contributed by these minerals proceeds fast. Moreover, because of their expansive character, the expansive additive can definitely improve the efficiency of self-healing.

However, technical problems of this approach have to be solved. For instance, if the minerals are directly added into the concrete mixtures without any protection, once they come in contact with water during the mixing of concrete, they immediately start to react [33]. As a result, these added minerals are consumed before cracking. Moreover, when an expansive additive is used, expansion always occurs in the interior of the concrete matrix, which could cause damage [37]. Therefore, when applying mineral admixtures to realize self-healing, pre-processing, like encapsulation, is necessary. Ahn and Kishi [38] tried to apply the encapsulation technique to store free lime (CaO) and anhydrite ($CaSO_4$) for self-healing before the mixing of concrete. One more vital requirement for this self-healing mechanism is that water should be continuously available in cracks, the same precondition as required for bacteria-based self-healing [32, 34, 35].

2.4 Autogenous self-healing

Autogenous self-healing in Portland cement concrete has attracted much attention since it was observed many years ago. According to Hearn [39], the phenomena of autogenous self-healing had already been noticed in water retaining structures, culverts and pipes by Hyde [40] by the end of nineteenth century. In 1920s, a more systematical analysis of autogenous self-healing was executed by Glanville [41]. After that, autogenous self-healing of cracks in bridges was also investigated [42, 43].

The effect of autogenous self-healing on water leaking through cracks was extensively studied by Clear [44], Hearn [45] and Edvardsen [46]. Moreover, Reinhardt et al. [47] correlated this effect with different temperatures and crack widths. The reduction of chloride ingression through cracks due to autogenous self-healing was reported by Fidjestol et al. [48], Ramm et al. [49] and Otsuki et al. [50]. Apart from the durability aspect, the improvement of mechanical properties of concrete due to autogenous self-healing has been explored as well. Lauer and Slate [51] demonstrated that the tensile strength measured perpendicular to the crack plane increased after autogenous self-healing of cracks. In that study, the influence of age and curing conditions were also taken into account. Similarly, the recovery in strength of concrete was also found by Dhir et al. [52] and Granger et al. [53].

Although the effects of autogenous self-healing have been investigated for many years, the results of the study at the components of reaction products formed the autogenous self-healing process are not consistent. Jacobsen and Sellevold [54] found some newly formed portlandite and ettringite in cracks in high performance concrete. Schlangen and Ter Heide [55] detected newly formed C-S-H in cracks after the cracked samples were cured in water. They concluded that autogenous self-healing was caused by further hydration of unhydrated cement clinker. Edvardsen [46] found calcium carbonate ($CaCO_3$) in cracks after autogenous self-healing. The investigations by Yang et al. [56] and Qian et al. [57] also evidenced the existence of $CaCO_3$ in cracks. When CO_2 in the air dissolves in water, CO_3^{2-} ions diffuse into cracks through the crack mouth [46]. $CaCO_3$ precipitates when the concentration of Ca^{2+} and CO_3^{2-} ions reach supersaturation level [46]. Moreover, Sisomphon [32] reported that, since the concentration of CO_3^{2-} nearby the crack mouth is higher than that inside the cracks, $CaCO_3$ tends to precipitate nearby the crack mouth.

2.5 Summaries on required conditions for various mechanisms of self-healing

The required conditions for the aforementioned mechanisms of self-healing are summarized in Table 1. As indicated by Table 1, different mechanisms of self-healing require different conditions. Therefore, self-healing mechanism can be designed for concrete structures under particular servicing conditions.

Table 1: Required conditions for different mechanisms of self-healing

Conditions / Mechanisms	Presence of water in cracks	Presence of CO_3^{2-} ions in cracks	Small crack width	Methods to supply healing agent to cracks
Self-healing based on adhesive agents	No. Cracks should be absent with water	No. It is not necessary.	No. Wide cracks can be healed depending on the amount of agents added.	It is necessary to supply the healing agent to cracks by using capsules or vascular systems.
Self-healing based on bacteria	Yes. Water should be continuously available.	Yes. CO_3^{2-} ions should be continuously available.	No. Cracks with a width of 450 µm can be healed [29].	It is necessary to immobilize bacteria before adding them into concrete.
Self-healing based on mineral admixtures	Yes. Water should be continuously available	No. It is not necessary.	No. Cracks with a width of 200 µm can be healed [35].	Healing agents, i.e. mineral admixtures, can be added into concrete directly or encapsulatedly.
Autogenous self-healing	Yes. Water should be continuously available.	No. It is not necessary. But presence of CO_3^{2-} ions facilitates the healing.	Yes. Cracks with a width less than 50 µm can be healed [58].	--

3. METHODS TO SUPPLY HEALING AGENTS TO CRACKS

Except autogenous self-healing, self-healing with other mechanisms usually requires the supply of healing agents to the cracks. By now, the main methods to supply healing agents to cracks *artificially* are by using capsules or vascular systems. The state-and-art of these two methods is presented below.

3.1 Encapsulation techniques

The encapsulation technique can be used to store and protect healing agents in the matrix. Once cracks appear and propagate through the matrix, some capsules can be hit and ruptured by the cracks, the healing agents are released into the cracks, provided that the healing agents are in liquid form. This concept has been used for self-healing in polymers for more than 10 years (see Figure 2) [59].

Similar to the case in polymer materials, many researchers have used capsules to supply liquid-based adhesive agents to cracks in cementitious materials for self-healing, as mentioned in Section 2.1. Also, solid healing agents, such as bacteria and mineral admixtures described in Section 2.2 and 2.3, respectively, can be protected by using capsules and mixed into concrete.

There are various encapsulation techniques available so far. They can be grouped into *physical* encapsulation and *chemical* encapsulation [60]. These techniques are widely used in the food industry and pharmaceutical industry [60]. Experience of encapsulation in the food industry and pharmaceutical industry can be useful for encapsulating healing agents for concrete mixtures.

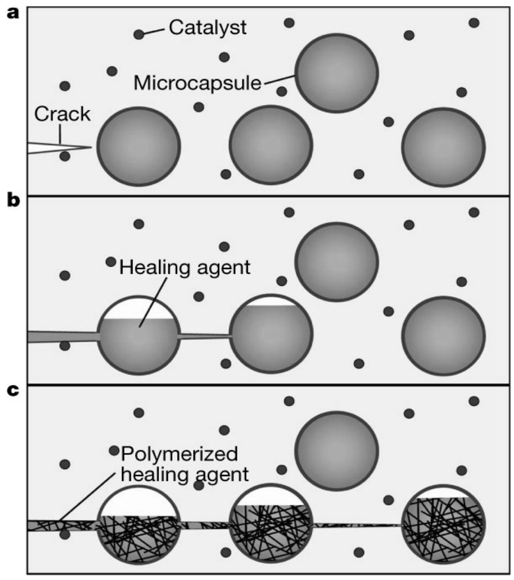

Figure 2: Material with microcapsules for self-healing [59]

In general, the use of capsules for self-healing has the advantage that self-healing of cracks can take place automatically [59]. However, there are some limitations of this method. Since the capsules are randomly dispersed in the matrix, only a small part of them can be hit and ruptured by cracks [61-63]. In order to guarantee that cracks are able to intersect the capsules, the strength of capsules is usually designed to be lower than the matrix itself. This, however, can lead to a lower overall strength of the material.

3.2 Technique of vascular systems

For delivering liquid healing agents to cracks also a vascular system can be applied. As shown in Figure 3, if cracks intersect any part of the vascular system inside the material, liquid healing agents can be transported to the cracks through the vascular system [64-67]. As a result, self-healing of cracks takes place.

Dry [68] embedded some pipes in concrete as a simple vascular system to supply healing agents to cracks. It was found that the capacity of self-healing was high when using this method [68]. Stimulated by these positive results, some researchers are now investigating methods to install vascular systems in concrete structures. For instance, Pareek et al. [69] set up vascular systems in concrete structures by creating canal networks. To do that, steel bars with smooth surface are pre-embedded inside the concrete matrix during casting. After the concrete is cured for 1 day, the embedded steel bars are pulled out. As a result, a canal is created inside the concrete matrix. A network is formed when these canals are connected with each other. Sangadji and Schlangen [70] are also investigating the installation of a vascular system in concrete structures. “Porous concrete” (see Figure 4) is developed for creating vascular systems. The pores inside the porous concrete are of high connectivity and the pore size is in the order of magnitude of millimeters. Similar to the canal networks proposed by Pareek et al. [69], when any part of the porous concrete network is intersected by cracks, liquid healing agents can be delivered to cracks via this system.

As discussed before, one of the disadvantages of the use of capsules is that only a limited amount of liquid healing agent can be supplied to cracks. In comparison, sufficient liquid agents can be delivered to cracks by using vascular systems, leading to high efficiency of self-healing. However, extra efforts are needed to inject the liquid healing agents into the vascular systems. Moreover, by now it is still difficult to install a vascular system inside the matrix and the vascular system is usually vulnerable.

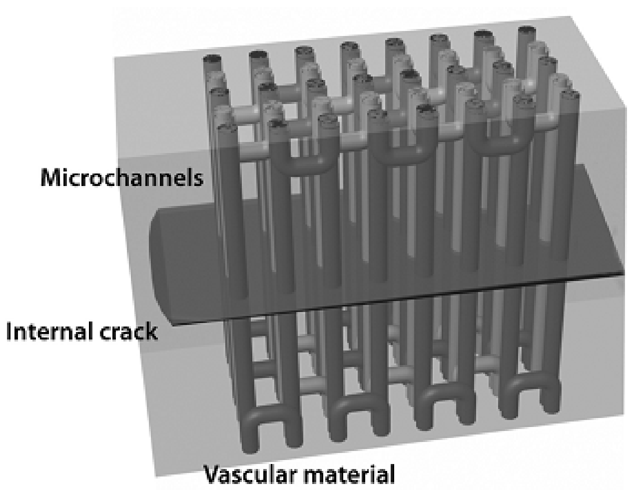

Figure 3: Material with a vascular system for self-healing [65]

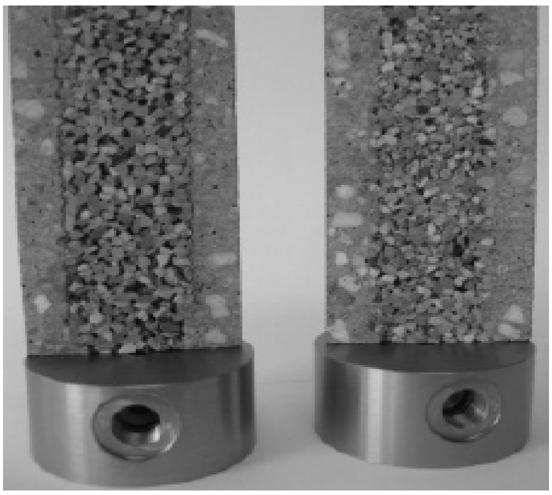

Figure 4: Porous concrete for vascular systems [70]

4. SERVING CONDITIONS OF REINFORCED CONCRETE STRUCTURES AND POTENTIALLY APPLIED SELF-HEALING MECHANISMS

Concrete structures serve under various conditions. As presented in Table 2, some concrete structures serve under water. As shown in Table 1, one of the most necessary conditions for autogenous self-healing and self-healing based on bacteria or mineral admixtures is that water is present in cracks. Therefore, for concrete structures under water autogenous self-healing and self-healing based on bacteria or mineral admixtures can be designed. However, self-healing based on adhesive agents is not suitable for concrete structures under water because presence of water in cracks prevents from the release of adhesive agents into cracks and the hardening of adhesive agents.

Table 2: Serving conditions of reinforced concrete structures and potentially applied self-healing mechanisms

Mechanism / Structures	Self-healing based on adhesive agents	Self-healing based on bacteria	Self-healing based on mineral admixtures	Autogenous self-healing
Concrete structures under water	hardly	recommended	recommended	recommended
Concrete structures under ground	For concrete in water-rich regions, it is hardly applied.	recommended	recommended	recommended
Concrete structures in the open air	recommended	Need to supply additional water	Need to supply additional water	Need to supply additional water
Indoor concrete structures elements	recommended	hardly	hardly	hardly

For concrete structures underground, in water-rich regions water can penetrate into cracks immediately after cracking. This case is similar to that concrete structures are under water. Autogenous self-healing and self-healing based on bacteria or mineral admixtures, but not adhesive agents, are recommended. In some regions where water is not extremely rich, it takes some time for water to fill the cracks. For this case self-healing based on adhesive agents is a possible mechanism to be applied. Moreover, wet-dry cycles usually take place in cracks of concrete structures underground. Wet-dry cycles facilitate penetration of CO_2 into cracks. As shown in Table 1, dissolved CO_2 facilitate the precipitation of calcite during autogenous healing and the healing based on bacteria.

In most of the serving time of concrete structures in the open air, water is absent in cracks. This condition makes autogenous self-healing and self-healing based on bacteria or mineral admixtures very difficult. As presented in Table 1, dry condition of cracks is good for self-healing based on adhesive agents. Therefore, self-healing based on adhesive agents can be applied in these structures. If autogenous self-healing and self-healing based on bacteria or mineral admixtures are designed for these structures, additional water should be supplied to cracks in purpose. Additional water can be supplied to cracks by using a vascular system described in Section 3 or by spraying water to concrete surfaces.

For indoor concrete structures elements, it is difficult to supply additional water to cracks. In this case, autogenous self-healing and self-healing based on bacteria or mineral admixtures are hardly applied.

5. CONCLUSIONS AND PROSPECTS

In this paper, different mechanisms of self-healing, i.e. self-healing based on adhesive agents, self-healing based on bacteria, self-healing based on autogenous self-healing were described. Their required conditions were summarized. The previous investigations showed that all mechanisms of self-healing are effective to some extend under particular conditions. However, there is still a lack of information about the efficiency of different self-healing in field application. Actually, concrete structures serve under particular conditions that can limit the choice of self-healing mechanisms. In this paper, concrete structures were categorized according to serving conditions. Potential self-healing mechanisms are pointed out according to the required conditions of each self-healing mechanism. However, no detailed information is available about the application of self-healing in concrete structures under particular serving conditions. More investigations of self-healing in concrete structures in field are necessary.

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