Self-Healing Concrete with Crystalline Admixture—A Review

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> Abstract: The scope of the present paper is to understand the effects of crystalline admixture on the selfhealing capacity of the cementitious composites. Previous studies were examined and a conclusion was drawn to the effect that different additives to crystalline admixture tend to improve self-healing of concrete for larger cracks. It is recommended that initial treatment with chemical admixture can stimulate and heal further cracks and it has the better repeatability trend in mixing with the concretes and mechanical recovery is possible even under repetitive preloading. Effective self- healing with chemical admixtures even under open-air exposure, leads to study the importance of a service ability design parameter including the maximum allowable crack width by repeatability analysis as a function of the exposure with the concept of sealable crack width.

Key words: crystalline admixture; permeability reducer admixture; cracking

1 Introduction

Concrete is used as construction material due to its fire resistance, high compressive strength and low expense. However concrete has small tensile strength $[1]$ and hence is prone to cracking .Cracks reduce anti-chloride-corrosion, anti-permeability and anti-carbonisation in a great way^[2]. This, in turn, reduces the durability and carrying capacity of structures and non-repair can lead to destruction and even collapse^[3].

Self-healing materials can autonomously heal cracks, improve service life with reasonable cost, and thereby improve the sustainability of the structure^[1,3] and guarantee for reinforcement in their entire life $[4,5]$. Concrete by itself has self-healing properties known as autogenous healing. In the past attempts have been made to induce autogenous healing process in cement such as the addition of Portland cement^[6-9], applications of microencapsulated healing agents $[10-13]$, the use of crystalline admixtures and bacterial concrete^[14].

ACI Committee has reported that there is a better adoption of self-healing properties in self-healing concrete when there is an addition of crystalline admixture (CA) also known as permeability reducer admixtures^[15]. Crystalline admixture materials react quickly with water and are used as hydrophilic materials. Calcium silicate hydrate (CSH) density is further enhanced which resists water penetration. In the presence of water, matrix components react with tricalcium silicate (C_3S) . Crystalline deposits can withstand pressure up to 14 bars just like hydrophobic materials. However, the results are a mixed bag. For instance, crystalline admixture had a higher rate of self-healing in small cracks (less than 0.05 mm) while they were inefficient in more extensive cracks $^{[16]}$. Combination of crystalline admixture and self-healing concrete achieved 400 μ m^[17] against 150 μ m when there was no mixture. Admixtures have been analysed in different exposures $[18,19]$ and methodologies based on mechanical strength, duration and recovery property that depend on the structure. Why cracks are formed are due to the expansion and shrinkage in concrete with temperature difference, applied force, structural mechanism, and dryness on the concrete surface. Fig.1 describes the different types of concrete cracks and their solutions are found in Table1. Fig.2 describes the self-healing process. The aim of this paper is to explore the crystalline admixture impacts on the self-healing capacity of concrete by either partially or entirely resealing the cracks and as in some cases to show recovery of mechanical properties. The study takes into account both high-performance and standard strength concrete fibre-reinforced cementitious concrete. The methods of the review are presented in Section 2. Section 3 critically reviews and interprets different studies used in the analysis. Sections 4 and 5 discuss and conclude the studies along with future recommendations.

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Fig.1 Classification of crack

2 Methods

The present review employes various standard sources for the literature search which includes scholarly web search engine such as Google Scholar and IEEE transaction and journals including Elsevier, Springer, and ACM and so on. In addition, numbers of manual searches were conducted within the review papers and individual documents' reference sections. A total of 22 different research studies matched the search criteria for this review.

Fig. 2 Classification of self-healing process in concrete by Meharie et al^[20]

3 Results

An extensive researches on SHC are listed in Table 2. Section 3.1 analyses the various healing materials, methods and results from the selected probes. The studies on self-healing concrete were based on admixture materials, crack type, different combinations of additives, different research approaches and different environmental exposures. In the initial stages , in order to offer better admixture to heal the concrete, different kinds of cracks and different admixtures were examined.

3.1 General description of the study

Examination on the basis of different environmen-

tal exposures to evaluate the best crystalline admixture is examined by Roig-Flores *et al*^[16], Roig-Flores *et* $a^{[21]}$, Roig-Flores *et al*^[23] and Reddy and Ravitheja^[22]. Examining the influence of crystalline admixture constitution on the basis of mechanical and physical properties was done by Joa *et al*^[26], Ferrara and Krelani^[24] and Reddy *et al*^[25]. The durability and waterproofing ability of concrete were examined by Pazderka and Hajkova^[27]. Other different additives that were added to the crystalline admixture and its effect on the self- healing process were examined by Sisomphon *et al*^[28,29] and Jaroenratanapirom and Sahamitmongkol^[30]. Further, the effect of self-healing with and without crystalline

admixtures was examined by Ferrara *et al* [31,32].

3.2 Self-healing capability with crystalline admixture in different environmental conditions

The life service of the building constructs is improved by adding self-healing materials which autonomously repair cracks and small damages. Furthermore, these types of materials could perform as a life saver for the future issues in the structure or expensive reparations. As reported by the ACI Committee, crystalline admixtures which are a type of permeability reducer admixture are characterized as hydrophilic in nature, which increases solubility in water. When mixed with water they form water-insoluble pore /crack blocking deposits that increases the concentration of calcium silicate hydrate (CSH) and resists water penetration. Controlled creation of specific damages (*e g*, cracks) and the evolution of such damages under different environment form the basis of a study of self-healing materials.

From the results presented in Fig.3 which are based on the analysis of the study Roig-Flores *et al*^[16] and Roig-Flores *et al*^[23], it may be inferred that there are better results with CA than with control concrete. However, CA natural self-healing character alone could not support to bring complete self-healing. Hence the mix design of control and CA concrete is shown in Table 3.

The crack closing under various environmental conditions and under different exposures such as water immersion(WI), wet or dry cycles (WD),water contact(WC) and air exposure (AE) under laboratory conditions were examined by Reddy and Ravitheja $[22]$ using the proportion of the crystalline admixture with 1.1 % weight of the cement.

The optic microscope was used to observe the physical closing of the crack and quantification was carried out through crack geometrical parameters. When water temperature is 30 ℃, the healing is complete, and the healing behaviour is dependent on the exposure and the presence of 4% of crystalline admixture. When disclosures concrete is mixed with 1.1% crystalline admixture, it regains the split tensile strength and compressive strength as control concrete durability. When cement was replaced with silica fume^[25] (5%, 10%, 15%) and with the addition of 1.1% crystalline admixture, all mixtures have a considerable amount of closing ability and strength regain capacity under different exposure conditions. Borg *et al*^[40] assessed the crack sealing capacity in chloride environments of different concrete mixtures, incorporating supplementary cementitious materials as well as self-healing enhancing crystalline admixtures. The result showed that immersion under sea water showed better crack healing performance compared to distilled water, which indicates that the higher the chloride content, the better and faster the healing.

3.3 Effect of crystalline admixture on physical and mechanical properties

Including crystalline admixtures in construction material is a novel idea. However, a thorough investigation is needed before they can be included in building material.

Joa *et al*^[26] examined the crystalline admixture of 3% by weight into cement and lime mortars. The modules of elasticity are found by dividing the stress and its corresponding strain . The mechanical properties of high performance fibre reinforced cementitious composites were examined by Ferrara and Krelani^[24]. Ferrara *et* $al^{[31,32]}$ attempted to investigate and characterize the crystalline admixtures impacts on the self-healing capacity of the concrete material by assessing their capacity to partially or completely heal cracks and mechanical properties. The interaction between the crystalline admixture performance and the dispersed fibre reinforcement is favourable in 'chemical pre-stressing' of the same reinforcement, from which the materials' mechanical performance recovery has be largely in-

Material	Cement	w/c ratio		Coarse aggregate Coarse aggregate $(20-12.5 \text{ mm})$ $(12.5-4.75 \text{ mm})$	Sand	Steel fiber	Crystalline (0.51%) admixture (1.1%)	Compaction factor
Control concrete	425.5	0.45	599	599	656 5	40	-	0.87
C A concrete	420.82	0.45	599	599	656.5	40	4.68	0.87

Table 4 Mix design of control and CA concrete

creased up to levels even greater than the virgin uncracked materials' performance.

3.4 Waterproofing ability and durability of concrete with crystalline admixture

Crystalline materials include surface-applied spraying or coating, integral waterproofing admixtures and sealing and repairing mortars. Mainly, the waterproofing admixture is applied for the latest buildings with the foundation structure from waterproof concrete. Most of the literature in the field of concrete reinforcement has been focused mainly on the durability and waterproofing ability of the concrete by crystalline admixtures. Regardless of these two properties, other properties have not been credibly established so far. The other characteristics include the influence of the lifetime of waterproofing by the crystalline admixture in the cementitious materials. Furthermore, water vapour permeability is one of the critical challenges due to the surface condensation. Pazderka and Hajko $va^{[27]}$ conducted various experimental measurements for durability and waterproofing ability of concrete using crystalline admixture. The waterproofing effect of crystalline material is obtained by the reaction of various chemical components incorporated in the concrete material. The crystalline admixture has the potential to reduce the water vapour permeability of concrete by 16%-20%. Furthermore, water pressure test was also conducted for a range of time intervals. The initial phase of cement hydration showed complete waterproofing ability of concrete with a crystalline admixture only after 12 days. The mechanical properties such as compressive strength of the prepared concrete materials are also examined. The compressive strength of the specimens from concrete without admixture and concrete with 2% crystalline admixture were almost the same after 28 days.

The analysis of water permeability and self-healing of a high performance concrete (HPC) was conducted by Escoffres *et al*^[33]. The potentials of HPC with fibers and crystalline admixture (HPFRC-CA) and HPC with fibers (HPFRC) were examined. The maximal crack widths is 39% lower and water permeability is 3.1 times lesser than that of HPC, under constant loading. The cracks of HPFRC-CA and HPFRC were

completely healed in comparison to 60% for those of the HPC, under a 7-day constant loading and a continuous water flow. HPFRC-CA showed slower self-healing kinetics than that of the HPFRC. However a higher load needs to be exposed in the HPFRC-CA to attain the initial permeability. The samples under SEM imagining showed the existence of ettringite and calcite in the HPFRC whereas the HPFRC-CA showed aragonite. Pazderka $[34]$ investigated the crystalline admixtures and their impact on water proofing properties of concrete. The experiment results revealed that crystalline admixtures have the ability to reduce the water vapour permeability value of concrete around 16%-20%. The complete waterproofing potential was attained only after 12 days of concrete creation

3.5 Self-healing concrete with crystalline admixture and other additives

In recent years, numbers of various cementitious additive materials have been analyzed in the field of self-healing concrete. The enhanced self-healing ability of concretes incorporating of calcium sulfoaluminate based expansive additive (CSA) and crystalline additive (CA) was conducted. The permeability and microstructure of cracked concretes were examined.

The concrete materials with crystalline additive (CA) and calcium sulfoaluminate based expansive additive (CSA) had a favourable surface crack closing ability^[28]. A ternary mixture consisting of a blend of Portland cement ,10% CSA and 1.5 wt% CA was an optimal mix design in which the rate of water passing dropped to zero in 28 days, and surface crack width up to 400 µm was completely closed. Investigations with crystalline additive (CA), calcium-sulfoaluminate based expansive additive (CSA) and ordinary Portland cement by Sisomphon and Copuroglu^[29] showed the repeatability of self-healing process were completely healed within 28 days. However, smaller cracks were sealed within 14 days. When Jaroenratanapirom and Sahamitmongkol $[30]$ evaluated the mortars constituting various additional cementitious materials such as fly ash, silica fume, and crystalline admixture. It was found that all mortars show self-healing capacity to some extent. For a crack range of 0-0.05 mm width, crystalline admixture (CA) showed the best self-healing performance. When the crack width was larger, silica fume became more effective in self-healing, showing that all mortars were crystal-like materials. Albertini^[35] examined the self-healing ability of concrete. The width of crack has a direct influence on the mechanical self-healing capacity of concrete. By increasing the width of crack, the capacity of self-repairing decreases. Self-healing has the effect of improving the durability of the material. The addition of crystalline admixture accelerates the reactions of self-healing and makes the bridges that crystals form between the crack sides stronger. Crystallizing agent increases both self-sealing and self-repairing properties. Compression enhances self-healing. A novel approach has been presented by Al-Kheetan *et* $al^{[35]}$ to add a dual-crystalline hydrophobic admixture in fresh concrete for improving hydrophobicity against chloride and harmful chemicals. The optimum performance was found in mixtures with 2% admixture. Mixture with 2% aqueous hydrophobic admixture revealed marginal strength compared to 2% crystalline cementitious hydrophobic admixture, although water protection appears to be better in cementitious mixture.

3.6 Self-healing of concrete with and without crystalline admixtures

Crystalline additives can also serve as self-healing engineering additions. Crystallization reactions can undergo a delayed activation whenever the material comes back into contact with water or environment moisture. It can happen after crack formation in later ages.

Ferrera *et al*^[31] added employed crystalline admixture comprising a mixture of cement, sand and active silica to the raw concrete constituents before mixing. Properties such as stiffness and load-bearing capacity have been estimated by means of 3-point bending tests. The results indicated that underwater immersion crack healed properly. The crystalline additive speed up the crack healing process. Air exposure was not enough to induce any significant recovery.

3.7 Repeatability analysis of self-healing concrete with and without crystalline admixtures

During construction, damage of infrastructure is frequent due to various overloading and changing environmental conditions, and therefore robustness of the self-healing mechanism should concerns in repeatability phenomenon.

Cuenca and Ferrara^[37] analyzes the repeatability of autogenous and engineered self-healing in fiber re-

inforced concrete (FRC) with and without crystalline admixtures. From the obtained results, the following conclusions can be drawn: The highest healing rate was reached for specimens immersed in water, followed by those subjected to dry/wet cycles and by those exposed to open air. For FRC with crystalline admixtures, a general trend was found for all exposure healing conditions in which the crack sealing and healing effectiveness after the $2nd$ healing was is higher than that after the 1st. It can be reasonably assumed that the crystalline admixture is able to provide a reservoir for delayed hydration reactions over time. This study presents the results from a comprehensive laboratory investigation of the application of 1%, 2% and 8% of crystallizing aqueous and cementitious hydrophobic mineral in fresh concrete. Despite the high slump in the fresh mixture, no segregation was observed in the matured concrete. There was a marginal reduction of strength when a high percentage of admixtures was used. Despite this, significant reduction of water absorption was observed indicating greater hydrophobicity. The optimum performance was found in mixtures with 2% admixture.

Cuenca and $Ferrara^{[41]}$ conducted a study to assess the crack healing effectiveness wherein the specimens were healed for one month and tested again up to a crack width of 0.25 mm (0.01 in.) (cycle after 1st healing). After that, specimens were healed for two months further (2nd healing) and finally, they were cracked once again up to 0.25 mm (0.01 in.). Regarding the repeatability, a general better trend was found for the mix with crystalline admixtures, in which, in addition, the maximum load regain was measured after the 2nd healing cycle rather than after the 1st healing. More recently, Cuenca *et al*^[38] attempted to investigate the repeated cracking and healing procedure for one year. The results show that, for the same healing period, the specimens immersed in water reached the largest crack closures. The result of the study showed that the crystalline admixture might favour long-term self-sealing capacity under repeated cracking and healing events.

4 Discussion

The studies shows that each research group applies their own methods of testing.

The present review suggests focusing on autonomous healing by the capsule and vascular-based materials for future research based on two reasons: First, the autogenous self-healing reliability is identified to be lower and secondly, they depend on the composition

of the cementitious matrix. Furthermore, these type of healing agents react on contact with air or moisture make them less interesting as premature curing of the agent inside the capsules, and the reaction will not start immediately after release.Reaction to heat is less interesting as it requires human intervention. Therefore the authors choose a second component in the cementitious matrix or a second component provided by additional capsules.

Based on the high performance fibre reinforced cementitious concrete and standard strength concrete, Ferrara and Krelani^[24] stated that the mechanical properties have significant effects with some internal chemical pre-stressing of the fibres triggering the expansive reaction of the admixture observed in HPFRCCs and a suitable self-healing indices to quantify the mechanical properties. Joa et $al^{[26]}$ studied the mechanical properties of self-healing admixtures exposing to porosity and water absorption capacity with crystalline admixture of 3% by weight. Reddy *et al*^[25] replaced cement with silica fume (5%, 10%, 15%) and with the addition of 1.1 % crystalline admixture indicating that all mixtures have a considerable amount of closing ability and strength regain capacity for different exposure conditions. Concrete with 10% silica fumes and 1.1% crystalline admixtures have a 100% strength retrieved capability and complete crack closing ability. Sisomphon *et al*^[28] used Calcium sulfoaluminate based expansive additive (CSA) and crystalline additive (CA) in cement-based materials and the mixtures had a favourable surface crack closing ability. The study revealed that water is very essential for self-healing of the cracks. The repeatability test is helpful to analyze the robustness of the self-healing mechanism, chemical admixtures have better repeatability trend in mixing with the concretes, and mechanical recovery is possible even under repetitive preloading.

5 Conclusions

Autogenous healing and free healing have been studied in the current work. The present study mainly focused on the autogenous healing capability of concrete materials with additive geo-materials, simulation and modelling of self-healing phenomenon, engineered cementitious composites' self- healing capacity and finally the conclusion for the admixtures mechanism on the efficiency of self-healing concrete. The review of the various literature revealed that intrinsic self-healing in cementitious materials is capable of extending their service-life. Self-healing concrete saves a lot for the construction industry by reducing the costs of repair and maintenance.

It is hoped by the authors of this work that the information that can be gleaned here may be useful for future researchers in this field. Presently, the application of capsule-based self- recovery systems seems to have promising desirable outcomes for the future construction industry due to its effective features to protection during mixing. However, the vascular healing and intrinsic healing potentials have limitations in movement, curing, abilities, workability and structural coherence.It appears that self-healing concrete is still in its nascent stage. Still, others believe that this type of concrete would be better suited for structures such as tunnels, bridges and highways rather than for houses and buildings. The improvements in the autogenous healing behaviour and mechanical reinforcement properties includes the post-tensioning of cementitious materials by shrinkable polymers which incorporate the nanoclay and polyvinyl alcohol (PVA) fibres using novel techniques. Autogenous healing concrete could optimise if free water that is nanoclay, a critical crack width, fibres control the cracks and right curing environment are available in the mixture. The intrinsic or natural character of cementitious materials has to be taken into explanation in scheming autogenous healing concrete, like a relatively high water-binder ratio and the low efficiency of rehydration of unhydrated cement nuclei. A promising solution to crack repair not only improves the long-term durability and service life, but also ingests the anthropogenic CO , emissions, which jeopardise the conditions of earth and is harmful to the life of humans.

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