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BIOLOGICALLY INDUCED SELF HEALING CONCRETE: A FUTURISTIC SOLUTION FOR CRACK REPAIR

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Abstract

Concrete is a mixture of cement, water, sand and other aggregates in adequate proportions. Its high tensile strength and ability to withstand a vast range of environmental changes makes it the first choice for construction material. One of the major problems associated with concrete is its permeability because penetration of gases and/or liquids from the surrounding environment into the concrete, followed by physical and/or chemical reactions within its internal structure/s leads to irreversible damages. Although cement has autonomous capacity to heal, however cracks <0.2mm width can only self-heal. Biomineralization is one of the best eco-friendly techniques to tackle the problem of cracks in concrete structures. Biologically induced self-healing is beneficial in addressing all the drawbacks of concrete matrix. The most promising technology for producing crack resistant/highly self healing concrete in near future seems to be "BacillaFilla": genetically modified version of *Bacillus subtilis*, is a "custom –designed" bacteria to embed deep into the cracks in concrete where they produce a mix of calcium carbonate and a special bacteria glue that hardens to the same strength as of the surrounding concrete.

Key words: Concrete, Self-healing, Bio-mineralization, Biologically induced self-healing, Cracks.

Introduction

Concrete is today's material of choice for construction world-wide because of its strength and cost effectiveness. Concrete is a mixture of cement, water, sand and other aggregates in adequate proportions. It has high tensile strength and can withstand vast range of environmental changes quite effectively. Properties such as strength, permeability, crack formation and corrosion properties define the overall quality of concrete mortar. Since concrete is composed of aggregates of various sizes connected with hydration products generated by mixing cement and water, cracks in concrete can occur at any stage of the service life due to volume instabilities such as autogenous shrinkage and/or drying shrinkage. Once cracking sets in anywhere in reinforced concrete member, not only the stiffness reduces but corrosion of supporting iron bars also occurs as a result of penetration of rain water and oxidizing substance (Mihashi and Nishiwaki, 2012). Many traditional methods are in use for crack repair like impregnation of cracks with epoxy based fillers, latex binding agents such as acrylic, polyvinyl acetate, butadiene styrene, etc. However, there are several disadvantages associated with these in vogue repair systems, namely i) different thermal expansion coefficient/s compared to concrete, ii) weak bonding,

iii) environmental & health hazards along with iv) high cost of the polymers/chemicals.

Although cement has autonomous capacity to heal but cracks smaller than 0.2mm width can only self-heal. Biomineralization is one of the best eco-friendly techniques to tackle the problem of cracks in concrete structures. Biomineralizationis defined as a biological process in which organism/s create a local microenvironment with conditions that allow optimal extracellular chemical precipitation of a mineral phase. Biomineralization processes involve calcium carbonate (CaCO₃) precipitation, which can occur by two different mechanisms: biologically controlled and/or induced (Lowenstan and Weiner, 1988). In biologically controlled mineralization, the growing organism controls the process to a large extent, i.e. nucleation and growth of the mineral particles. The organism synthesizes minerals in a form that is unique to that species and independent of environmental conditions. Examples of controlled mineralization are magnetite formation in magneto-tactic bacteria (Bazylinski et al., 2007) and silica deposition in the unicellular algae and diatoms, respectively (Barabesi et al., 2007). The calcium carbonate production by bacteria is generally regarded as "induced" as the type of mineral produced is largely dependent on the environmental conditions

and no specialized structures or specific molecular mechanism are thought to be involved (Rivadeneyra *et al.*, 1994). The technology of calcium carbonate deposition or microbial concrete is called as "Microbially Induced Carbonate Precipitation" (MICCP) (Dhami *et al.*, 2012). This review article aims to provide an overview about the problem of cracks in concrete, biologically induced self-healing solutions using microbes with its associated advantages and challenges.

Self-Healing Property of Cement

There are many drawbacks associated with concrete production and structures as well. It is estimated that cement production alone contributes 7% to global anthropogenic carbon dioxide emissions, due to the sintering of limestone and clay at a temperature of 1500 degree Celsius. So, from an environmental viewpoint, concrete does not appear to be a sustainable building material (Jonkers et al., 2010). Permeability of concrete poses a major problem because penetration of gases and/or liquids from the surrounding environment. The ensuing physical and/or chemical reactions within internal structures of concrete lead to irreversible damage (Vahabi et al., 2012) as this causes corrosion in the embedded metal structures used to reinforce the building. Cracks occur in concrete due to various mechanisms such as shrinkage, freeze-thaw reactions and mechanical compressive and tensile forces. Although micro cracks (width smaller than 0.2 mm) do not affect strength properties of concrete structures directly but they contribute to material porosity and permeability which leads to ingression of aggressive chemicals such as chlorides, sulfates and acids that results in concrete matrix degradation and embedded premature corrosion of the iron reinforcement hampering the structures durability in the long term (Jonkers and Schlangen, 2008).

Several studies have shown that concrete structures have a certain capacity of autonomous healing of micro cracks (Neville, 2002; Reinhardt and Jooss, 2003; Li and Yang, 2007; Worrell *et al.*, 2001). The actual capacity of micro crack healing is primarily related to the composition of the concrete mixture. In the autonomous self-healing process dissolution and reprecipitation of calcium carbonate occurs. The crack-penetrating water would not only dissolve calcite (CaCO₃) particles present in the mortar matrix but would also react together with atmospheric carbon dioxide with partially hydrated lime constituent such as calcium oxide and calcium hydroxide according to the following reactions:

$$Ca (OH)_2 + CO_2 \longrightarrow CaCO_3 + H_2O$$
(2)

Biologically induced self-healing or MICCP is beneficial in addressing all the above drawbacks of concrete matrix. The bacteria induce precipitation in between the pores of the cement matrix thereby reducing the pore size and consequently increasing the strength and decreasing the permeability at the same time.

Mechanism of Biologically Induced Self-Healing

Calcium carbonate precipitation is governed by four key factors: (i) calcium (Ca $^{2+}$) concentration, (ii) concentration of dissolved inorganic carbon (DIC), (iii) pH and (iv) availability of nucleation sites (Ariyanti *et al.*, 2011; Muynck, 2010; De Belie, 2010). The primary role of microorganism in carbonate precipitation is mainly due to their ability to create alkaline environment (high pH and [DIC] increase) through their various physiological activities (Hammes and Verstraete, 2002).

Three/four main groups of microorganism that can induce calcium precipitation are: (i) photosynthetic microorganism such as cyanobacteria and microalgae; (ii) sulphate reducing bacteria (iii) some species involved in nitrogen cycle and iv) lactate metabolism. The MICCP phenomenon appearing in aquatic environment is caused by photosynthetic microorganisms. Photosynthetic microorganisms use CO_2 in their metabolic process (eqn. 3) which is in equilibrium with HCO_3^{-1} and CO_3^{-2} as described in eq. 4 given below. Carbon dioxide consumption by photosynthetic microorganisms shifts the equilibrium resulting in increase in pH (eqn. 5) (Ariyanti et al., 2011). When this reaction occurs in the presence of calcium ion in the system, calcium carbonate is produced as described at chemical reaction in eqn.6.

$CO_2 + H_2O -$	\rightarrow (CH ₂ O) + O ₂	(3)
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 $2\text{HCO}_{3}^{-} \longleftrightarrow \text{CO}_{2} + \text{CO}_{3}^{2-} + \text{H}_{2}\text{O}$ (4)

$$\text{CO}_3^{2-} + \text{H}_2\text{O} \iff \text{HCO}_3^{-} + \text{OH}$$
 (5)

$$Ca^{2+} + HCO_3^- + OH^- \iff CaCO_3 + 2H_2O$$
 (6)

Another pathway uses sulphur reducing bacteria (SRB). Abiotic dissolution of gypsum (CaSO₄.H₂O) (eqn. 7) produces a system rich in sulfate and calcium ions. In the presence of organic matter and absence of oxygen, SRB reduces sulphate to H_2S and HCO_3^- as described in eqn. 8. When the H_2S degasses from the environment, pH of system will increase and precipitation of calcium carbonate ensues (Ariyanti *et al.*, 2011).

$$CaSO_4 + H_2O \longrightarrow Ca2^+ + SO_42^- + 2H_2O$$
(7)
2(CH_2O) + SO_4^{2^-} \longrightarrow HS^- + HCO_3^- + CO_2 + H_2O (8)

Urease enzyme activity in microorganisms has been used as a tool to induce precipitation of calcium carbonate (Bekheet and Syrett, 1977; Mc.Connaughey, 2000). Hydrolysis of urea by urease enzyme in heterotrophic microorganism produces carbonate ions and ammonium. This mechanism results in higher pH and

$$CaO + H_2O \longrightarrow Ca (OH)_2$$
(1)

enrichment of carbonate ion (Hammes and Verstraete, 2002). One mole of urea is hydrolyzed intra-cellularly to one mole of ammonia and one mole of carbamate (eqn. 9), which spontaneously hydrolyses to one mole of ammonia and one mole carbonic acid (eqn. 10). Ammonia and carbamate subsequently equilibrate in water to form bicarbonate and 2 moles of ammonium and hydroxide ions as described in eqn. 11 and 12.

 $CO (NH_2)_2 + H_2O \longrightarrow H_2COOH + NH_3 \qquad (9)$

 $NH_2COOH + H_2O \rightarrow NH_3 + H_2CO_3$ (10)

 $2NH_3 + 2H_2O \longrightarrow 2NH^{4+} + 2OH^{-}$ (11)

 $2OH^{-} + H2CO_{3} \rightarrow CO_{3}^{2-} + 2H_{2}O$ (12)

Overall reaction:

 $CO (NH_2)_2 + 2 H_2O \longrightarrow 2NH^{4+} + CO_3^{2-}$ (13)

The presence of calcium ions in the system leads to calcium carbonate precipitation once a certain level of super-saturation is reached. The Calcium Carbonate precipitation mechanism induced by urease activity is illustrated in Fig1. (adapted from Hammes and Verstrate, 2002)

History and current status of biological induced mineralization:

For the bacteria to mineralize there are certain properties it should possess. Firstly, the bacteria should be resistant to high pH (alkali resistant). Secondly, the bacteria should be able to produce copious amounts of minerals needed to plug or seal the freshly formed crack. Since concrete structures are designed to last at least 50 to 100 years, bacteria should remain viable for a long period of time. Therefore a specific group of alkaliphilic spore-forming bacteria were selected to be used in biological induced mineralization. Several species from the genus Bacillus have been used for incorporation into concrete. Thirdly, as the concrete matrix is oxygenic due to ingress oxygen (diffusion through matrix capillaries) incorporated bacteria also need to be oxygen brilliant (Jonkars et al., 2010). Fourthly, the bacteria should be resistant to high calcium ion concentration and should produce calcium carbonate at greater amounts. Fifthly, it should be able to withstand high pressure conditions as if embedded inside the matrix and should remain viable for long periods of time. Lastly, the bacteria used should be non-pathogenic.

Furthermore, Urea is an important organic nitrogen carrier in natural environment and is commonly used as an agricultural fertilizer (Nieslen *et al.*, 1998). Several research groups have explored free as well as immobilized microorganisms for inducing self healing capability in concrete, which have been summarized in Table 1.

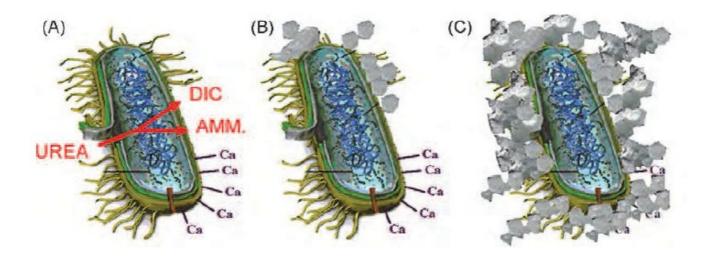


Fig 1: Illustration of calcium Carbonate precipitation mechanism induced by urease enzyme activity in microorganism

(A) Ca ions attracted to bacterial cell. Urea is added, dissolved carbon (DIC) and Ammonia (AMM) are released into environment. (B) The whole cell is encapsulated; (C) Calcium Carbonate precipitation ensues.

S. No	Microorganism Used	Proposed Mechanism	Major finding	Author (Year)
1	Bacillis pasteurii, Pseudomonas aerugiinosa	Urease metabolism	 Used as sealant increased Compressive Growth inhibited in high alkaline environment High efficiency in shallow cracks. <i>P. aerugiinosa</i> contributions were insignificant. 	Ramachandran et al., 2001
2	Bacillis pasteurii	Poly Urethane (PU) immobilized, Urease metabolism	 Cells retained high metabolic rate. Protected from adverse environment. No change in rate of metabolism due to immobilization. Accumulation of calcite in deeper cracks. 	Bang <i>et al.</i> , 2001
3	-	Poly Urethane immobilized urease enzyme	• Rate of precipitation lower than free enzyme.	Bachmeiri et al., 2002
4	Bacillus sphaericus	Silica sol incorporated Urease metabolism	 Incorporation with calcium source. Effective in sealing cracks of 0.01mm-0.6mm. Reduced concrete permeability 	De Belie and De Muynck., 2008
5	Bacillus cohnii, Bacillus halodurans, Bacillus pseudofirmus	Two component self- healing system	 Substrate and microbes added into the concrete. Formation of 100µm sized calcite particles, superior in filling large cracks. 	Jonkers and Schlangen., 2008
6	Bacillus sphaericus	Encapsulation of bacteria, nutrient and PU in glass rod	 Higher the concentration of microbe more was the precipitation. PU contributed to increase in strength. 	Wang <i>et al.</i> , 2010
7	Bacillus pseudofirmus and B.cohnii	Two component Healing system ; Bacteria and calcium lactate	 20-80µm sized mineral-like precipitate on crack surface. Functionality limited to young cement. Decrease in nitrogen loading in environment. 	Jonkars <i>et al.</i> , 2010
8	Microalgae (Chlorella)	Photosynthetic Pathway	 Renewable source Easily cultivatable No optimization done yet 	Ariyanti et al., 2011

Table 1: Different Bacterial populations and their potential for bio mineralization

Challenges Ahead

There are commercial products available based on traditional methods used for crack repair like CPR # 400-A binding paste, Thompsons emergency MASTIC, DAP but no product is yet available based on biologically induced self-healing. Biologically induced self-healing is a promising technology to increase the strength, life and durability of concrete structures. But there are several challenges to implement this in the practical world. Applying the bacterial mixture externally not only increases the cost but also needs manual inspection at regular intervals. The major challenge faced is about keeping the cells viable for a longer duration of time till the concrete structure is present. It has been found that bacterial spores are viable in young concrete but with time the microbial viability decreases. Moreover, the bacteria should acts as healing agent innumerable times once it has been incorporated. This can only be possible when new spores are generated once the crack is repaired. The impregnation of microbes in PU or sol-gel offers a solution to increase the viability but optimization of this has yet to be accomplished. Moreover, using microalgae and other microorganisms for crack remediation has as yet not been fully investigated.

Future Prospects

Promising results on the use of microorganisms for the improvement of durability of building materials have drawn the attention of research groups all over the world. It is clear that the work done by several research groups, focusing on different materials, can only improve our understanding on the possibilities and limitations of biotechnological applications on building materials. Investigations are still going on the retention of nutrient and metabolic products, as they have an influence on survival, growth and biofilm formation. The most promising technology for producing crack resistant/highly self healing concrete in near future seems to be "BacillaFilla": is a genetically modified version of *Bacillus subtilis*. It has been "custom-designed to burrow deep into the cracks in concrete where they produce a mix of calcium carbonate and a special bacteria glue that hardens to the same strength of the surrounding concrete" developed at Researchers at the University of Newcastle in the UK (Dilow, 2010).

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