“SELF HEALING CONCRETE – THE NEED”

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ABSTRACT

Crack formation in concrete is common, but a typical phenomenon related to durability. Percolation of cracks may lead to leakage problems or ingress of deleterious materials, causing deterioration of the concrete matrix or corrosion of embedded steel reinforcement. Durability can be enhanced by preventing further ingress of water and other substances. In recent years a bacteria-based self-healing concrete is being developed to extend the service life. A two component healing agent is added to the concrete mixture. The agent consists of bacteria and an organic mineral precursor compound. Whenever cracks occur and water is present the bacteria become active and convert the incorporated organic compounds into calcium carbonate, which precipitates and is able to seal and block cracks. This paper aims to review the development of bacteria-based self-healing concrete, introducing the proposed healing system. Different stages in the development are discussed, and some recommendations for further research are given.

I INTRODUCTION

Concrete in most structures is designed to crack in order to let embedded steel reinforcement take over tensile stresses. Crack formation is also a typical phenomenon related to durability. Percolated cracks may lead to leakage problems or ingress of harmful materials, which can cause deterioration of the concrete matrix or reinforcement corrosion. Durability can be enhanced by preventing further ingress of water and other substances. About 10 billion tons of concrete is produced every year and out of that about 4-5% of the concrete produced gets broken down or collapses due to the formation of crack. So it is very important to find a solution for this problem, and self-healing concrete seems to be a solution that has been much needed.

Self-healing is characterized by regaining performance after a defect occurs. Damage targeted in bacteria-based self-healing concrete particularly relates to increased durability and leakage prevention and extending service life of concrete structures. Jonkers (2007) introduced a two-component healing agent to be added to the concrete mixture, consisting of bacteria and a mineral precursor compound. Upon cracking the system is activated by ingress water. Bacteria convert the mineral precursor compound into the mineral calcium carbonate, better known as limestone. Precipitation of the limestone on the crack surface enables sealing and plugging of the cracks, making the matrix less accessible to water and other deleterious materials.

In the laboratory a fully functional bacteria-based self-healing system exists, which will be introduced in this paper. New studies will focus on further development of the system in order to make practical application of the material feasible.

II CONCEPT

A self-healing material that is capable of reaching its original state (SHC) that has been observed over time (autogenic) has been observed in many old structures which have remained standing for long periods of time in spite of the fact that they have limited maintenance. This observation concludes that the cracks heal when moisture interacts with non-hydrated cement clinker in the crack. Nevertheless, in present-day constructions the cement is lowered as a result of modern construction methods. Hence, the amount of available non-hydrated cement is therefore reduced.

Self-healing concrete is a product that will biologically produce limestone to heal cracks that appear on the surface of concrete structures. Specially selected types of the bacteria genus Bacillus, along with a calcium-based nutrient known as calcium lactate, and nitrogen and phosphorus, are added to the ingredients of the concrete mixture when it is being mixed. These self-healing agents can lie dormant within the concrete for up to 200 years. However, when a concrete structure is damaged and water starts to seep through the cracks that appear, the spores of the bacteria germinate on contact with the water and nutrients. Having been activated, the bacteria start to feed on the calcium lactate. As the bacteria feed, oxygen is consumed and the soluble calcium lactate is converted to insoluble limestone.
solidifies on the cracked surface, thereby sealing it up. The consumption of oxygen during the bacterial conversion of calcium lactate to limestone has an additional advantage. Oxygen is an essential element in the process of corrosion of steel and when the bacterial activity has consumed it all it increases the durability of steel reinforced concrete constructions. The two self-healing agent parts (the bacterial spores and the calcium lactate-based nutrients) are introduced to the concrete within separate expanded clay pellets 2-4 mm wide, which ensure that the agents will not be activated during the cement-mixing process. Only when cracks open up the pellets and incoming water brings the calcium lactate into contact with the bacteria do these become activated. Testing has shown that when water seeps into the concrete; the bacteria germinate and multiply quickly. They convert the nutrients into limestone within seven days in the laboratory. Outside, in lower temperatures, the process takes several weeks.

Figure: this shows how the crack is healed over the period of time.
1.1. Self-healing

Current material design in engineering follows the concept of damage prevention. An alternative design principle is that of self-healing materials, according to the concept of damage management as introduced by Van der Zwaag (2007). Damage formation does not necessarily cause problems, if it is subsequently healed in an autonomous process. Self-healing materials have to serve some roles and meet several properties. Damages should be sensed, followed by transportation of healing agent to the damage site, triggering repair of the damage. In the ideal case self-healing materials are cheap and have properties equal or superior to currently used materials, with the ability to heal defects of any size, multiple times, completely and autonomously. In case of concrete durability performance is mainly considered for damage to be healed, in order to reduce costs of repair and maintenance. An overview of characteristics for self-healing concrete is given by Jonkers (2007). Target for self-healing concrete is to reduce matrix permeability by sealing or blocking cracks.

Healing agent is incorporated in the concrete matrix and acts without human intervention.

Preference lies in agents working as a catalyst, enabling multiple healing events. To make the material technically and economically competitive, healing agent should be cheap in relation to the low price of concrete, remain potentially active for long periods of time and be concrete compatible to not negatively affect material characteristics.

1.2. Microbial healing

Concrete already has a built-in healing mechanism due to on-going chemical, physical and mechanical processes. Most significant is precipitation of calcium carbonate (Edvardsen 1999). Average limit for which healing can still occur is a crack width of 0.2 mm. Carbonation reaction lies at the base of the calcium carbonate production, where diffused carbon dioxide reacts with the hydration product calcium hydroxide as can be seen in Equation 1.

\[ \text{EQ. (1):} \]
\[ \text{CO}_2 + \text{Ca(OH)}_2 \rightarrow \text{CaCO}_3 + \text{H}_2\text{O} \] (1)

The principle of microbial healing also lies in the precipitation of calcium carbonate (Jonkers et al. 2010). Ingress water activates dormant bacteria. Dense layers of calcium carbonate are produced by bacterial conversion of an incorporated mineral precursor compound. In case of calcium lactate the reaction is as given in Equation 2, where bacteria only act as a catalyst.

\[ \text{EQN. (2):} \]
\[ \text{Ca(C}_3\text{H}_5\text{O}_2 \) + 7\text{O}_2 \rightarrow \text{CaCO}_3 + 5\text{CO}_2 + 5\text{H}_2\text{O} \] (2)

From the metabolic conversion of calcium lactate carbon dioxide is produced, which further reacts with the calcium hydroxide from the concrete matrix according to the chemical reaction in Equation 1, producing additional calcium carbonate. Massive production of large, over 100 μm sized (Van der Zwaag et al. 2009), crystalline calcium carbonate precipitates seal and block cracks, preventing further ingress of water and possible other substances that may attack the concrete matrix or embedded reinforcement.
Metabolic pathways for bacterial influenced carbonate production are diverse. Several researchers selected hydrolysis of urea as a suitable pathway for bio cementation (e.g. de Belie and de Muynck 2008, Ramachandran et al. 2001). During the ureolytic induced carbonate formation the surroundings are alkalinized, favoring the precipitation of calcium carbonate in the form of calcite (Dick et al. 2006). In the overall reaction one mole of urea is hydrolyzed into one mole of carbonate and two moles of ammonium ions. Jonkers (2007) deliberately chose a metabolic pathway based on organic calcium salts utilization instead of hydrolysis of urea to prevent possible detrimental effects on the concrete matrix or embedded reinforcement when produced ammonia is further oxidized to nitric acid by bacteria.

III WHAT BACTERIA IS USED FOR PREPARATION OF SHC

Bacteria are relatively simple, single celled organisms. The bacteria used were Bacillus Pasteurii, Bacillus Cohnii, and Bacillus Filla etc. It is a bacterium with the ability to precipitate calcium carbonate in the presence of any carbonate source. The bacterium is used for the same and for the improvement in the strengths of the concrete test specimens were observed. The microbes are a bacillus species and are completely not harmful to human beings. They precipitate inorganic crystals hence the healing of the cracks takes place in the concrete and it can withstand any temperature conditions.

- Most of the Bacteria belongs to genus bacillus are fulfilling the required criteria discussed above.
The suitable chemical precursor found to be most suitable is “calcium lactate” \( \text{Ca(C3H5O2)2} \).

Fig. Bacteria used in the preparation of self-healing concrete.

(a) B. Pasteurii (b) B. Cohnii

(c) B. Filla
IV HEALING AGENT

1. Direct addition

Healing agent mainly consists of bacteria and a mineral precursor compound. First important consideration was to choose concrete compatible bacteria. Bacteria should survive and remain active in the highly alkaline environment. 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 was selected. The thick cell-walled spores are produced by bacteria when living conditions become less favorable. Spores are characterized by resistance to high mechanical and chemical stress (Sagripanti and Bonifacino 1996) and have extremely long life spans in dormant state, for some species up to 200 years (Schlegel 1993). When conditions are suitable spores germinate and transform into active vegetative bacteria, namely in alkaline surroundings with access to water and a food source. Several species were selected from the genus Bacillus for concrete incorporation (Jonkers 2007). Tests on concrete compatibility showed no significant influence on flexural and compressive strength characteristics for concentrations of added bacteria up to $10^9$ cm$^{-3}$. 

Figure: Healing agents
Special interest lies in the effect of incorporated mineral precursor compounds on concrete properties. Majority of healing agent consists of the organic mineral precursor compound which is by the bacteria metabolically converted to carbonate ions which subsequently precipitate with calcium ions in form of limestone on the crack surface. Several organic precursor materials such as specific amino acids appeared suitable candidates as this hardly affected concrete compressive strength (Jonkers 2007). Calcium lactate however, appeared to be the most suitable compound as its application as main healing agent ingredient resulted in even enhanced concrete compressive strength values (Jonkers and Schlangen 2009). The combination of suitable bacteria and calcium lactate as mineral precursor compound calcium lactate indeed resulted in production of calcium carbonate precipitates in concrete cracks. The observed mineral production in time however appeared limited when calcium lactate and bacterial spores were directly added in unprotected form to the concrete mixture, probably due to full integration of the precursor compound in the matrix limiting its access to bacteria (Jonkers and Schlangen 2009). Also viability of bacterial spores appeared limited to 2–4 months by direct addition. This is likely due to continuing reduction in pore size of the cement paste by further cement hydration. Mercury Intrusion Porosimetry (MIP) shows that in time pore diameters come below 1 μm, the average size of Bacillus spores (Jonkers et al. 2010).

Increased potential for long-term viability and activity may be reached when integrated bacterial spores are immobilized or protected and the precursor compound is kept accessible for bacterial conversion. Opted solution is encapsulation of the two-compound healing agent in a protective reservoir (Jonkers and Schlangen 2009, Jonkers et al. 2010).

Figure: Scenario of crack-healing by concrete-immobilized bacteria (Jonkers 2007). Ingress water activates bacteria on fresh crack surfaces, bacteria start to multiply and precipitate calcium carbonate, which eventually seal and plug the crack and protect embedded steel reinforcement from further external attack.
2. Encapsulation LWA

In order to substantially increase functionality in time, the incorporated two component healing agent was protected by immobilization porous expanded clay particles (Wiktor and Jonkers 2011). Impregnation of the light weight aggregates (LWA) occurred under vacuum, twice with a calcium lactate solution and finished with a bacterial spore suspension. LWA were dried in an oven in between impregnation treatments and before application. After completion of the impregnation treatments expanded clay particles contained 6% healing agent by weight.

In order to make a concrete mixture self-healing, part of the aggregate material in the range of 2–4 mm was replaced by similarly sized healing agent containing LWA, corresponding to a healing agent content of 15 kg m\(^{-3}\) concrete (Jonkers 2011). Replacement of a significant fraction of sand and gravel for LWA changed then material into light weight concrete, affecting material characteristics such as a substantial reduction in compressive strength. Capacity to heal cracks was substantially improved for concrete containing in LWA encapsulated healing agent (Jonkers 2011, Wiktor and Jonkers 2011). Viability of incorporated bacterial spores increased to 6 months, with experiments on-going.

V PREPARATION OF BACTERIAL SOLUTION

Primarily 12.5g of Nutrient broth (media) is added to a 500ml conical flask containing distilled water. It is then covered with a thick cotton plug and is made air tight with paper and rubber band. It is then sterilized using a cooker for about 10-20 minutes. Now the solution is free from any contaminants and the solution is clear orange in color before the addition of the bacteria.

Later the flasks are opened up and an exactly 1ml of the bacterium is added to the sterilized flask and is kept in a shaker at a speed of 150-200 rpm overnight. After 24 hours the bacterial solution was found to be whitish yellow turbid solution.

Figure: Solution without bacteria (only media)
VI  COMPRESSION  STRENGTH  TEST

The cubical Molds of size 150mm x 150mm x 150mm were cleaned and checked against the joint movement. A coat of oil was applied on the inner surface of the Molds and kept ready for the concreting operation. Meanwhile the required quantities if cement, fine aggregate and coarse aggregate (passing through IS sieve of 20 mm size and retained on 4.75 mm) for the particular mix are weighed accurately for concreting. Fine aggregate and cement were mixed thoroughly in a hand mixer such that the color of the mixture is uniform. Then, weighed quantity of coarse aggregate is added to the mixer and then it rotated till uniform dry mixture is obtained. Then, calculated quantity of bacterial solution and water was added and mixing was continued for about 3 to 5 minutes to get a uniform mix. The wet concrete is now poured into the Molds and for every 2 to 3 layers and compacted manually. After concreting operations, the upper surface is leveled and finished with a mason’s trowel. The corresponding identification marks were labeled over the finished surface and they were tested for 7 and 28 day strengths in a compressive strength testing machine.

Figure: Solution with Bacteria
Figure: Concrete cube subjected to compression

How the healing agents are applied:

- **By direct application**
  
The bacteria and the chemical precursor (calcium lactate) are added directly while making the concrete.

- **Encapsulation LAW**
  
The part of the coarse aggregate is replaced by the light weight aggregate (LWA), which is impregnated with twice the calcium lactate solution and the spores of bacteria.

  After impregnation the clay particles with 6% healing agents and the concrete is made.
The following Figures show how the crack has been fulfilled:

Figure: Crack develop in concrete

Figure: Cure by bacteria phase 1

Figure: Fully cure by bacteria phase 2
VII EVALUATION

According to De Muynck et al. (2010) significant added value for self-healing materials can be expected in case of substantially reduced need for manual inspection and repair. In a recent publication Wiktor and Jonkers (2011) quantified the crack-healing capacity of concrete containing LWA encapsulated self-healing agent. Maximum crack width for full healing was found to be 0.46 mm for bacteria-based specimens, what was significantly higher than the 0.18 mm found for control specimens.

Activity of the bacterial system with LWA was shown by oxygen consumption measurements as oxygen is required for the metabolic conversion of calcium lactate (Wiktor and Jonkers 2011). Production of calcium carbonate was supported by microscopic inspection, Energy Dispersive X-ray (EDAX) analyses and measurements on Fourier-Transform Infrared (FT-IR) spectra, showing formation of calcite and aragonite. Concrete crack-healing in specimens containing LWA encapsulated healing agent was also functionally tested by permeability measurements which showed complete healing of cracks within a two week healing period (Jonkers 2011).

Concrete properties should not be negatively affected by addition of a healing agent to the concrete mixture. However, as was shown in aforementioned studies, incorporation of a large amount of bacteria and certain mineral precursor compounds like calcium lactate do substantially influence concrete compressive and tensile strength when the healing agent is added to the concrete mixture in larger volumes in form of LWA encapsulated healing agent. Optimization of healing agent characteristics is therefore still needed to decrease its effects on concrete strength reduction.

- The Concrete properties should not be negatively affected by addition of a healing agent to the concrete mixture.
- A large amount of bacteria and certain mineral precursor compounds like calcium lactate do substantially influence concrete compressive and tensile strength
- When the healing agent is added, Optimization of healing agent is necessary so that it won’t affect the characteristics of concrete

VIII CONSIDERATIONS

In order to consider practical application several characteristics have to be determined. Viability and functionality of incorporated bacteria is enhanced until several months after concrete casting. For practice long-term self-healing capacity is needed, ideally for the duration of the service life of the concrete structure. Also multiple healing events should be possible. At the moment full healing of cracks is accomplished for crack widths until about 500 μm. For acceptable appearance maximum crack width allowed in practice is 0.4 mm as stated in Euro code 2 (NEN 2005). As the values are promising, crack formation larger than 0.5 mm is common in practice, mainly compromising durability.

For practical applications possibility for localized plugging or sealing off surfaces of cracks with larger widths is recommended, preventing penetration of substances into the concrete matrix, extending the need for repair and maintenance.

Cost efficiency is also important. Concrete is a relatively cheap construction material, and adding a self-healing material to the concrete mixture has to be economically feasible. E.g. the return on investment price could come from savings on otherwise needed repair and maintenance costs. In order to minimize the price of the healing agent, its production should be straightforward with large output and little loss, minimizing the use of complex procedures, heating and cooling.

Also efficiency of the healing agent is an important factor. E.g. the above described system that uses LWA as a protective reservoir for the healing agent only contains 6% of healing agent by weight (Wiktor and Jonkers 2011). While in this specific case the self-healing capacity of concrete is significantly improved, its compressive strength is concomitantly reduced, limiting possibility for application to constructions in which leakage proofing and high strength is preferred. Therefore, development of a more efficient and economical healing agent could substantially widen the range of potential applications.
IX  FUTURE PERSPECTIVES

Currently a fully functional bacteria-based self-healing concrete system using LWA as storage reservoir is available on the laboratory scale. On-going studies in our laboratory investigate the possibility to use this system in practical applications.

A next step towards widening application possibilities is the development of a more efficient and economical agent that does not negatively affect concrete strength properties. Possibility for easy application and production on industrial scale at low costs should be considered. Next to healing capacity, long-term behavior and improvement of durability characteristics of the bacteria-based self-healing concrete material need to be determined, such as resistance to chloride penetration and freeze-thaw cycles. Long-term monitoring of larger scale experiments executed in the outdoors environment may reveal material behavior in practice. Feasibility of implementing the material in the market should then finally be determined by a full cost-benefit analysis.

Figure: a block showing cross section of a self-healing concrete
X  SUMMARY

The goal of this paper is to introduce bacteria-based self-healing concrete, currently being developed in the Micro lab of TU Delft. On the lab-scale a fully functional system exists. To the concrete mixture a healing agent is added, consisting of two components immobilized in expanded clay particles. Due to bacterial activity a calcium carbonate layer is deposited on the crack surface, sealing and blocking entrance to deteriorating substances. Further research and development is needed in order to make the material ready for application in practice. The system currently available may limit the field of application. Addition of a substantial quantity of light weight aggregates not only affects material properties, it can also impose economic restraints. Since potential advantages are mainly anticipated in reduction of costs for maintenance and repair and service life extension of concrete structures, the self-healing material needs to be cost efficient and durable.

XI  REFERENCES


