O U R N A L O F

Ceramic Processing Research

Crack self-healing technology based on bacteria

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Concrete cracks due to several reasons and through the cracks oxygen, water, chlorides and other aggressive agents can penetrate, decreasing the concrete durability, performance and life span. Even though concrete has autogenic capacity to heal the cracks, the size of the cracks that can undergo autogenous healing depends on the exposure conditions and remains below 0.6 mm. In order to improve this healing capacity Microbial Induced Precipitation (MIP) has been investigated. This method, which involves the precipitation of calcium carbonate (CaCO₃) induced by bacteria, was implemented at Delft University of Technology for the development of self-healing concrete for new structures as well as for mortar and liquid-based systems for repair of existing concrete structures. This paper presents an overview of the materials, their properties and the successful applications in the Netherlands and overseas.

Key words: Concrete, Crack, Durability, Self-healing, Bacteria.

Introduction

Crack healing in concrete

Crack formation in concrete is a common phenomenon. Cracks may occur because of shrinkage, thermal expansion, loading stress or creep. Even though cracks do not necessarily indicate a structural problem, they are considered undesirable. Even fine cracks can allow the ingress of oxygen, water, chlorides and other aggressive agents that may lead to premature corrosion of the steel reinforcement. This would lead to the decrease of durability, performance and life span of the concrete. Cracks are also undesirable due to aesthetic reasons.

Concrete has the autogenous capacity of healing cracks [1-3]. The recovery against an environmental action can be considered as healing if the width of a through-crack diminishes with time [1]. However the maximum crack width which could undergo autogenous healing was, in several studies, estimated to be below 0.17 mm [4-8]. Recent research has shown the potential sealing of cracks up to 0.59 mm of concrete submerged in artificial sea water [8]. The most significant factor that contributes to self-healing of cracks is the precipitation of calcium carbonate. In order to increase the crack-healing potential of concrete a number of specific agents can be incorporated to the concrete mix, such as superabsorbent polymers [9, 14], expansive agents and geomaterials [10, 11], and encapsulated liquid agents [12, 13]. Another approach

to improve the healing capacity is by means of Microbial Induced Precipitation (MIP). This method involves the precipitation of calcium carbonate induced by bacteria. Following this concept different building materials have been developed at the Delft University of Technology. This paper presents an overview over these materials: self-healing concrete for new structures [15] and repair systems for existing concrete structures. The repair systems are namely a mortar for patch repair [7, 16] and a liquid-based system for sealing of cracks and decrease of porosity [18, 19].

Bacteria as a healing agent

The production of calcium carbonate by bacteria, Microbial Induced Precipitation (MIP), is considered as an "induced" process because the characteristics of the minerals produced highly depend on the environmental conditions and so far no specialized structure are thought to be involved [1].

Different systems could contribute to MIP. The most commonly used are degradation of urea or bacterial ureolysis (enzymatic hydrolysis of urea) in a calcium rich environment. This process has been extensively studied because is easy to control and the output of calcium carbonate is considerable [20-22]. Urea is decomposed by bacteria with the aid of urease (bacterial enzyme) which results in the production of ammonium (NH₄⁺), dissolved inorganic carbon and an increase in pH which favors the precipitation of CaCO₃ [22]. The use of this system generates problems such as the environmental nitrogen loading due to the production of ammonia or negative effects to the concrete due to chemical reactions with ammonium salt [21].

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Another system considered for MIP is the use of bacterial mediated calcite precipitation by means of metabolic conversion of calcium lactate instead of urea hydrolysis. This is the system considered in this paper: alkaliphilic (alkali-resistant) spore-forming bacteria and calcium lactate as a nutrient source for the bacteria. Bacteria from genus *Bacillus* and more specifically related to the species *B.cohnii* [23], originally isolated from alkaline soil samples were chosen [24]. These bacteria are capable of healing cracks by direct and indirect calcium carbonate (CaCO₃) formation [24, 25]. The direct precipitation is due to the bacterial metabolic conversion of calcium lactate according to the following reaction:

$$CaC_6H_{10}O_6 + 6O_2 \rightarrow CaCO_3 + 5CO_2 + 5H_2O \tag{1}$$

The indirect formation of calcium carbonate is due to the reaction of metabolically produced (by the bacteria) CO_2 molecules with Ca (OH)₂ minerals (portlandite) that are present in the cement matrix, according to:

$$5CO_2 + 5Ca(OH)_2 \rightarrow 5CaCO_3 + H_2O \tag{2}$$

The reaction expressed in Equation 2 is homologous to carbonation, a slow process that naturally occurs in concrete due to inward diffusion of atmospheric carbon dioxide. This process can be substantially enhanced by the metabolic conversion of calcium lactate [24] with the production of in total 6 calcium carbonates equivalents from direct and indirect precipitation.

Bacteria to be used in concrete as a self-healing agent should be able to resist the concrete matrix incorporation and then be able to remain dormant in the hardened concrete until cracks appear. At that moment the bacteria is expected to be able to precipitate bio-minerals compatible with the cement matrix in the cracks filling them in order to heal or seal the cracks. At the same time the incorporation of bacteria and the above mentioned calcium lactate (as an organic nutrient source for the bacteria) should not negatively affect concrete characteristics such as workability, setting time, compressive strength among others. Bacteria that can resist concrete matrix incorporation exist in nature, and these appear related to a specialized group of alkali-resistant spore-forming bacteria [24]. Spores are specialized spherical thick-walled cells somewhat homologous to plant seed. These spores are viable but dormant cells and can withstand mechanical and chemical stresses and remain in dry state viable for periods over 50 years [25].

Self-healing Concrete with Bacteria

Laboratory scale testing

Initially spores of alkali-resistant bacteria related to the genus *Bacillus* were added to the concrete mixture [24]. The spores germinated after activation by ingress of water (through the cracks) and produced copious amounts of calcium carbonate-based minerals that precipitated in the cracks and filled them. However, in that study it was found that the self-healing potential was limited to young concrete (7 days cured), as viability and related activity of bacterial spores that were embedded directly into the concrete mix was restricted to about two months. The decrease in lifetime is due to the continuing hydration of the cement matrix resulting in matrix pore-diameter widths typically much smaller than the 1-mm sized bacterial spores.

In order to increase the lifetime of the bacteria incorporated in concrete it is necessary to immobilize both bacteria and organic compound (calcium lactate). A suitable carrier for the bacteria should survive the concrete mixing and later during the hardened stage of concrete break upon cracking and release the healing agent.

Tests were done by impregnating the healing agents into porous expanded clay particles with size range 1 to 4 mm [25]. Mortar samples were prepared in which 50% of the total aggregate volume was replaced with the expanded clay particles. This high percentage of replacement with particles that have a light weight and lower strength than the other aggregates lead to a decrease in compressive strength at 28 days when compared to mortar without replacement. However, the self-healing capacity of mortar with expanded clays impregnated with healing agent substantially improved when compared to mortar samples with nonimpregnated expanded clays. The healing capacity was assessed by permeability tests and light microscopy images in the study from Jonkers (2011).

Field application in Ecuador

The first field application of self-healing concrete took place on July 2014 in the Andean highlands in Ecuador. In the past lustrum, several canals that are part of the irrigation system in the province of Tungurahua in Ecuador got a concrete lining. Traditionally these canals were made by digging in the compressed soil. Because of infiltration of water through the walls (soil) the yield was very low. The newly built concrete linings soon started cracking, resulting in a large amount of leakage putting in danger the sustainability of the system. For a community economy based on agriculture, which depends for its water supply on these canals, a failure represented a big threat. Self-healing concrete seemed as a viable option to improve the sustainability and performance of the irrigation system.

Prior to the field application a concrete mix was designed taking into account the materials available in Ecuador, the strength and performance demands for the concrete linings and the building practice in the



Fig. 1. Impregnation on-site of expanded clays with bacterial spores and organic compound as food for the bacteria.



Fig. 2. Canal section casted with self-healing concrete (with bacteria) under working conditions only 5 days after casting.

highlands in Ecuador. The concrete mix included gravel with maximum size of 10 mm, sand, expanded clays with and without healing agent and natural fibres. The use of fibres in concrete increases the tensile capacity and assures a controlled crack width [26]. A fibre indigenous to Ecuador, Abaca, was chosen since this fibre has been successfully studied in Ecuador as reinforcement for mortar to improve the structural behaviour of houses under seismic forces [27].

The compressive strength, as tested in the laboratory, was 30 MPa for the mix with healing agent and 26 MP for the mix without healing agent [28]. The self-healing capacity of the concrete mixes was also evaluated in the laboratory.

In Ecuador, a section of canal that did not have concrete lining was chosen for the field application. This portion of canal is located at about 2900 meters above sea level. The local farmers, owners and users of the canal, clean the canal section from dirt and vegetation. They prepared the formwork and contributed with the sand and gravel from local extraction. The concrete mix designs were adjusted to the properties of the materials available in the site. The expanded clays were impregnated with bacteria and nutrients in the field (Fig. 1).

Three linear meters of concrete linings were cast with bacteria and 3 meters without bacteria. 110 litres of concrete were prepared at the time. The temperature during the casting was around 5 $^{\circ}$ C.

Three days after casting the formwork was taken off and two days later the water flow through the canal was reopened (Fig. 2). After 5 months (December 2014) the concrete linings show no signs of cracking or deterioration as was reported from the last inspection.

Mortar with Bacteria for Patch Repair

Laboratory scale testing

Most of the durability-related problems of patch repair systems are due to the lack of compatibility between the repair material and the concrete substrate. Strain-hardening cement-based composites (SHCC) have been studied as repair materials for concrete, for applications such as overlays in pavements or patch repair [5, 16]. SHCC are designed to have a large strain capacity possible because of the use of a low percentage of randomly distributed polymer fibres. Repair materials are subjected to differential shrinkage deformations which lead to tensile stress development. As a repair material SHCC can carry more tensile load and accommodate larger tensile strain than other repair systems. The tensile strain will translate into microcracking. In order to improve the healing capacity of SHCC a bacteria-based healing agent will be included into the mix.

Bacteria spores related to the species *B.cohnii* and calcium lactate as the food source were immobilized inside lightweight aggregates. For the repair mortar a different type of porous aggregate was chosen instead of the expanded clays used for concrete mixes. This decision was based on the particle sizes of both types of aggregates and on the advantages of angular shapes and good bonding with the concrete mix exhibited by the lightweight aggregates.

The mortar mix consists of cement, fly ash, limestone powder, lightweight aggregates with the healing agent, PVA fibres, water and superplasticizer.

The average compressive strength at 28 days of the SHCC with bacteria was 39.8 MPa and without bacteria 38.5 MPa. Under bending stress both materials behaved ductile and developed multiple cracks prior to failure. The mortar with healing agent showed reduced delamination with the concrete substrate, sufficient bond strength and improved flexural behavior as a composite (together with the concrete substrate) compared to the mortar without healing agent [16].

The healing capacity was evaluated by means of precracking under bending stress and then unloading the samples, curing them under water and testing them to failure. The results of these tests were compared to the

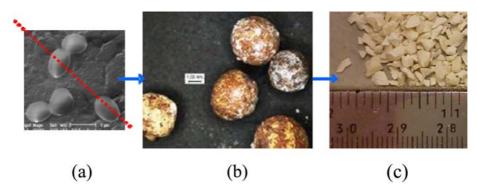


Fig. 3. (a) Bacteria mixed directly in concrete, short lifetime, (b) Impregnated expanded clays or other lightweight aggregates, viable but not efficient, (c) Compressed and coated pills, 100% healing agent.



Fig. 4. Self-healing mortar applied under dry conditions: (a) fresh state, (b) after 4 months.

results of tests done with samples that had the same curing conditions but no pre-cracking. As reported by the authors (2014a) the healed samples (with and without bacteria) have a higher strength than the reference samples (no pre-cracking). The results were slightly higher for SHCC with healing agent. Microscopic observation of the cracks after healing period showed calcium carbonate precipitates in all samples. Oxygen consumption measurements showed bacterial activity in the SHCC with healing agent but the lack of enhanced CaCO₃ precipitation could be attributed to limited amounts of feed applied.

A more efficient way to incorporate the healing agent into cement-based materials is to prepare particles which are 100% bacteria and food (Fig. 3). These new compressed particles need to be coated in order to survive the wet mixing process. Currently, tests are being done with 15 gr/ litre of mortar in combination with the lightweight aggregates. The mechanical properties of the material, healing capacity and behaviour as a repair material are being investigated.

Field applications in the Netherlands

During 2013 and 2014 the mortar for concrete repair described in section 3.1 was applied as patch repair in different locations in the Netherlands, under very diverse weather condition. The first application took place during May 2013 on a garage exposed to the weather elements in the East region of the Netherlands. The day of casting the temperatures were around 19 °C and during the previous 7 days only 1.3 mm of rainfall were register hence the concrete was dry. In the concrete structure to be repaired, the steel rebar already shown signs of corrosion that lead to the spalling of the concrete. The mortar was placed to completely cover the steel rebar and even the concrete surface (Fig. 4(a)). After 4 months the site was inspected and no signs of cracks were seen. After 1.5 years the patch repair is in good conditions without deterioration (Fig. 4(b)).

A completely different situation leads to the second field application. In an underground parking garage the retaining walls have had water leaking problems for months. After trying with different commercially available repair solutions, the self-healing mortar was applied. Traces of previous repair materials were removed with chisel, including traces of bitumen (Fig. 5(a)). The mortar was cast in place and no curing compound was applied (Fig. 5b). A month after the casting the repair looks sound. A very small leakage was still spotted but it has not lead to spalling even after 6 months (Fig. 5c).

An application that involved a higher volume of applied mortar took place in October 2014 in a tunnel that was built in the late 1930's. The concrete slab show signs of steel bar corrosion and concrete spalling. The concrete was removed in a patch of about $250 \times 500 \times 110$ mm. This patch was deep enough to expose the steel rebar which were cleaned from traces of

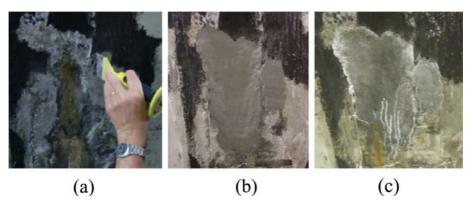


Fig. 5. Self-healing mortar applied under moist conditions: (a) Surface preparations, (b) Application, (c) A month later, small leakage in the bottom end of the application.

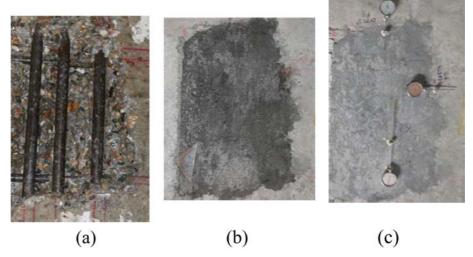


Fig. 6. Self-healing mortar applied at a tunnel: a) Surface preparations, b) Application, c) Monitoring the possible shrinkage and delamination.

corrosion (Fig. 6(a)). The mortar was mixed on the site and cast into the patch (figure 6(b)). The patch was covered with a geotextile material for the first 7 days. The possible cracking or delamination due to restrained shrinkage was monitored during the following two months (figure 6(c)). No delamination occurred and only very small shrinkage cracks were observed.

Liquid-based System for Concrete Repair

Laboratory scale tests

Wiktor and Jonkers (2012) implemented the principles of MIP to the development of a liquid-based system that aims at the sealing of cracks add decrease of porosity in aged concrete structures. The aim is achieved by means of the production of a calcium-based bio-mineral.

The system is composed of two solutions: Solution A which contains sodium-silicate (as an alkaline buffer), sodium gluconate (as a carbon source for bacteria growth) and alkaliphilic bacteria; and solution B, which contains calcium-nitrate (nitrate source for denitrification and calcium for CaCO₃ precipitation) and alkaliphilic bacteria (Wiktor & Jonkers 2015). The silicate-based

compound assures the formation of a gel inside the crack. This gel allows a rapid sealing of the crack and provides an optimum environment for bacteria to precipitate $CaCO_3$.

This liquid-based system was also tested as a healing agent injected into a porous network concrete (PNC). This type of concrete has a porous core which can be used as a media to transport healing agent to a fracture zone in a structure [29] as shown in Fig. 7. During the tests a crack was formed by means of bending stress and then the solution was injected through the porous network until it reached and flew out through the crack opening [30]. Even though mechanical regain in terms of strength and stiffness of bacteria-based post-healing beams was quite limited, crack sealing was observed and liquid tightness was assured.

Field applications in the Netherlands

In 2012 the first field application of the liquid-based system took place. A recently built structure in Breda showed multiple micro-cracks and during a rainy period water leaked into the building. The system described in section 4.1 was applied as shown in Fig. 8.

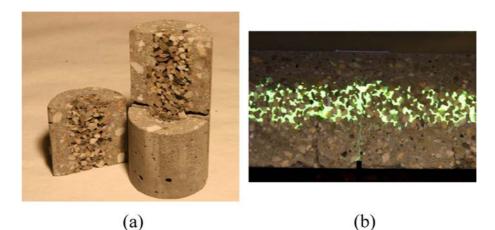


Fig. 7. Porous network concrete: (a) Concrete inspired by bone structure, (b) a healing agent can flow though the network and fill the cracks.



Fig. 8. Cracked concrete structure and crack sealing after impregnating with liquid-based repair system.

Within days the leakage of water dramatically decreased. A few months later core samples were taken to analyse the apparent crack sealing.

After this successful field application, in the recent years the impregnation system has been applied in ten other locations in the Netherlands, mainly in parking garages. One notable advantage of this system is the short reacting time, the application is done within few hours and the garage can be reopen. The efficiency of the liquid-based system to seal cracks was evaluated on-site by means of permeability tests. As reported by Wiktor & Jonkers (2015) the cracks that were treated exhibited only a few localized dripping spots or no leaking at all (Fig. 9(b)).

The liquid-based system was also applied to reduce the porosity of aged concrete and, in this way, improve the resistance to frost salt scaling (Wiktor & Jonkers 2015).

Conclusions

This paper presents an overview of the research done at Delft University of Technology to improve the crack healing capacity of concrete and other cement-based materials by Microbial Induced Precipitation (MIP). The system studied in Delft use bacterial mediated calcite precipitation by means of metabolic conversion of calcium lactate instead of urea hydrolysis. The principle was used to design self-healing concrete for new structures and repair materials for aged structures. Once the principle was probed in the laboratory several field applications has taken place in the past 3 years.

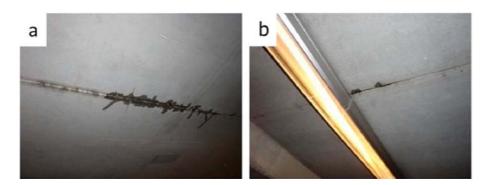


Fig. 9. (a) Leakage in untreated cracks, (b) reduced leakage through cracks after sealing them with liquid-based system.

The repair materials (with bacteria), both a liquidbased system and a mortar, have improved behaviour as a repair solution and has been successfully applied in the field in the Netherlands.

Self-healing concrete with bacteria and natural fibres was applied in Ecuador in 2014. This is the first field application of such a material. After six month the concrete shows no signs of cracks or deterioration.

Further research is taking place to improve the efficiency of the healing agent under numerous weather conditions and chemical attacks.

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