



El presente material comenzó a ser estudiado sistemáticamente en el año 2006 por el Microbiólogo Jenk Jonkers, Profesor de la Universidad Tecnológica de Delft (Holanda), en el marco de un conjunto de líneas de investigación que buscan caminos aptos para que el hormigón, ya colado en una estructura, se autorrepare. Es decir, que sea capaz de ir rellenando las fisuras que en él se producen a medida que se van presentando

1. Idea básica

La base del material es un hormigón normal al que se agrega un tipo de bacterias cuyas esporas, al ponerse en contacto con humedad, se activan y absorben y metabolizan el alimento que se ha introducido a ese fin en el hormigón, inician un proceso químico cuya culminación es la producción de un material que está en condiciones de sellar las fisuras por las que ha entrado la humedad que las activó.

2. Tipo de bacteria

En el presente caso se eligieron bacterias del género bacillus, que prosperan en condiciones alcalinas, como es el medio que impera en el hormigón endurecido y producen esporas: células esféricas de paredes delgadas que no deforman la estructura de la bacteria. Estas esporas, en vida latente siempre que estén secas, pueden permanecer estables frente a tensiones mecánicas o químicas por períodos de más de cincuenta (50) años. El tamaño de las esporas es del orden del micrómetro (μm).

3. Proceso biológico

Alimentando las esporas, en el momento en que se activan cuando se produce la fisura e ingresa en ella humedad, con lactato de calcio se logra que, al metabolizarlo, ellas combinen el calcio con iones de carbonato, que se encuentran en el ambiente, para formar carbonato de calcio (calcita), piedra caliza que se deposita en las paredes internas de las fisuras, o grietas, sellándolas.

4. Procedimiento tecnológico

Se coloca el lactato de calcio junto con las esporas en soportes que, llegado el caso, permiten el acceso del agua. A partir de ello las esporas germinan y se reproducen. Este soporte puede ser un material poroso, como son las arcillas expandidas, cápsulas de plásticos solubles biodegradables, etc.

En el presente trabajo son utilizadas arcillas expandida del tipo de las que se emplean como agregado grueso en los hormigones livianos, que tienen formas esféricas. El valor del diámetro de la acá utilizadas está comprendido entre 2 y 4 (mm). Con ellas se sustituyen el 50% del agregado de tales dimensiones.

Antes de ser utilizadas, las esferitas de arcilla se secan hasta peso constante (una semana a 40°C). La cantidad de arcilla expandida que se coloca por decímetro cúbico de hormigón, es la necesaria para incorporar a la masa 5×10^7 esporas y 15 (g) de lactato de calcio. La arcilla expandida se agrega al hormigón en el momento en que este se termina de elaborar. Esta sustitución de parte del agregado grueso pétreo por arcilla expandida, disminuye la resistencia a compresión del hormigón en el orden del 50% de la obtenida ensayando probetas cilíndricas a 28 días de edad del hormigón de base (sin sustitución de parte del agregado pétreo).

Cuando las fisuras comienzan a formarse en el hormigón, penetra en ellas agua que, al introducirse en los poros de la arcilla expandida germina las esporas. Las esporas, al germinar, se multiplican y se alimentan del lactato de calcio expresamente colocado. Al metabolizar el lactato de calcio, se inician un proceso durante el cual, finalmente, se combina el calcio con iones de carbonato y forman calcita, que es el material con el que se sellan las fisuras.

5. Características prácticas del material

Puede auto-sellar grietas de hasta 8 (mm), mucho mayores que las admisibles en

cualquier estructura de hormigón. Su costo es mayor que el de los hormigones comunes, esta diferencia se puede estimar en un orden del 40%, pero hay casos en los que, sin embargo, resulta una buena solución, incluso económica, como son los de reservorios y conductos de agua.

El ancho de fisura a partir del cual se desencadena el proceso de autorreparación es del orden de 0,15 (mm)

Dicho proceso se inicia en cuanto penetra agua o humedad del exterior en las fisuras. El tiempo necesario para sellar una fisura fue del orden de dos (2) meses en la presente serie de ensayos.

6. Proceso químico que culmina en el sellado de las fisuras

Si se ensayan dos hormigones similares, ambos con sustitución de parte del agregado grueso por arcilla expandida, pero uno con esporas y su alimento y el otro sin ello, en ambos se inicia un proceso de sellado de fisuras pero este solo eficiente en el que contiene esporas. Lo que ocurre se puede ser explicado de la forma que se detalla a continuación.

6.1) Hormigón de comparación (sin esporas):

Las partículas de cemento no hidratadas que siempre existen en las paredes de las fisuras recién formadas, se hidratan.

Además, las partículas de hidróxido de calcio contenidas en las paredes interiores de las fisuras, debido a su relativamente alta solubilidad, captan prácticamente todo el dióxido de carbono disponible en el agua que ingresó a las fisuras, formando carbonato de calcio

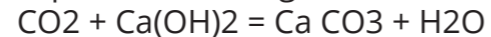
El carbonato de calcio precipita en las paredes internas de las fisuras, que es donde se encuentra el dióxido de calcio.

El proceso sigue de la siguiente forma: el hidróxido de calcio remanente en el hormigón se disuelve y difunde en la masa de agua interna a la fisura, reaccionando con el dióxido de carbono presente en las inmediaciones de las paredes internas de

ella. Este proceso químico conduce a la producción, y precipitación, de grandes cantidades del mucho menos soluble carbonato de calcio

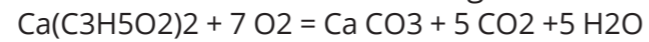
La cantidad de carbonato de calcio producida en el hormigón sin esporas no es mucha debido a la poca cantidad de dióxido de carbono presente en la limitada cantidad de agua que se ubica en el interior de la fisura. En resumen, falta dióxido de carbono para que la reacción continúe y se capte todo el hidróxido de calcio disponible en las caras de las fisuras

El proceso es el siguiente:



6.2) Hormigón autorreparable

En el hormigón autorreparable la reacción anterior se transforma en la siguiente:



Este proceso conduce a la precipitación de cantidades sustancialmente mayores de carbonato de calcio que en el caso anterior, debido a que este es producido no solo por la conversión del lactato en carbonato sino también, indirectamente, por la reacción química del (CO₂) producido metabólicamente por las esporas

En este caso, el dióxido de carbono producido por las esporas en las superficies interiores de las fisuras no se disuelve ni difunde sino que reacciona directamente con las partículas de hidróxido de calcio, allí presentes, para producir el carbonato de calcio adicional

En este segundo proceso, originado por la presencia de esporas y su alimento, se produce, en total, seis (6) moléculas de carbonato de calcio equivalente

Este sustancial incremento en la producción de carbonato de calcio es el que origina el sellado completo de fisuras y grietas indicado precedentemente.

Comentario: todas las estructuras de hormigón armado se fisuran en sus zonas traccionadas, o al menos esto es lo que debe suponer el Proyectista. Estas fisuras inevitables

que se producen en el hormigón, son aceptables si su apertura, en la superficie externa del cuerpo de hormigón, no supera valores del orden de 0,1; 0,2 ó 0,3 (mm) según la obra o la parte de ella de que se trate. Para saber si existen fisuras estructuralmente excesivas en una estructura de hormigón, hay que desarrollar un bien planificado Plan de Inspecciones Periódicas. Cuando se emplea hormigón autorreparable, si bien las visitas periódicas de Inspección no se van a eliminar, si se eliminará de ellas el tedioso trabajo de buscarlas. Y busca y encontrar fisuras excesivas, sobre todo en las zonas de difícil acceso, como por ejemplo la parte inferior de los tableros de puentes grandes y medianos es una tarea difícil y compleja. Este es un rubro importante cuando se habla del costo de una obra de hormigón, en el cual el costo particular del material empleado no es, ni mucho menos, la única variable a considerar y, muchas veces, ni siquiera la más significativa.

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BACTERIA-BASED SELF-HEALING CONCRETE

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A typical durability-related phenomenon in many concrete constructions is crack formation.

While larger cracks hamper structural integrity, also smaller sub-millimeter sized cracks may result in durability problems as particularly connected cracks increase matrix permeability.

Ingress water and chemicals can cause premature matrix degradation and corrosion of embedded steel reinforcement. As regular manual maintenance and repair of concrete constructions is costly and in some cases not at all possible, inclusion of an autonomous self-healing repair mechanism would be highly beneficial as it could both reduce maintenance and increase material durability. Therefore, within the Delft Centre for Materials at the Delft University of Technology, the functionality of various self healing additives is investigated in order to develop a new generation of self-healing concretes. In the present study the crack healing capacity of a specific bio-chemical additive, consisting of a mixture of viable but dormant bacteria and organic compounds packed in porous expanded clay particles, was investigated. Microscopic techniques in combination with permeability tests revealed that complete healing of cracks occurred in bac-

terial concrete and only partly in control concrete. The mechanism of crack healing in bacterial concrete presumably occurs through metabolic conversion of calcium lactate to calcium carbonate what results in crack-sealing. This bio-chemically mediated process resulted in efficient sealing of sub-millimeter sized (0.15 mm width) cracks. It is expected that further development of this new type of self-healing concrete will result in a more durable and moreover sustainable concrete which will be particularly suited for applications in wet environments where reinforcement corrosion tends to impede durability of traditional concrete constructions.

Keywords: Concrete crack-healing, permeability, bacteria, calcium carbonate formation

INTRODUCTION

Crack formation in concrete is a phenomenon that can hardly be completely avoided due to for example shrinkage reactions of setting concrete and tensile stresses occurring in set structures. While larger cracks can potentially hamper a structure's integrity and therefore require repair actions, smaller cracks typically with a crack width smaller than 0.2 mm are generally considered unproblematic [1-2]. Although such

micro cracks do not affect strength properties of structures they do on the other hand contribute to material porosity and permeability. Ingress of aggressive chemicals such as chlorides, sulfates and acids may result on the longer term in concrete matrix degradation and premature corrosion of the embedded steel reinforcement and thus hamper the structures' durability on the long term.

In several studies indications have been found that concrete structures have a certain capacity for autonomous healing of such micro cracks [2-5]. The actual capacity of micro crack healing appears primarily related to the composition of the concrete mixtures. Particularly mixtures based on a high binder content show remarkable crack-healing properties [5] what is due to delayed (secondary) hydration of matrix embedded non-hydrated cement and binder particles upon reaction with crack ingress water. Autogenous self-healing of cracks in traditional but also high-binder content mixtures appear limited to cracks with a width smaller than 0.2 mm [2-5]. This somewhat limited effectiveness appears largely due to the restricted expansive potential of the small non-hydrated cement particles lying exposed at the crack surface. Another limitation to application of high-binder content mixtures solely for the purpose of increasing self-healing capacities are recurrent policies which advocate sparse use of cement in concrete for sustainability reasons as current cement production contributes about 7% to global anthropogenic CO₂ emissions [6]. For latter reasons, alternative and more sustainable self-healing mechanisms are therefore wanted. One possible mechanism is currently being investigated and developed in several laboratories, i.e. a technique based on the application of mineral-producing bacteria. E.g. efficient sealing of surface cracks by mineral precipitation was observed when bacteria-based mixtures were sprayed or applied onto damaged

surfaces or manually inserted into cracks [7-13]. As in those studies bacteria were manually and externally applied to existing structures, this mode of repair can not be categorized as truly self healing. In several follow up studies therefore, the possibility to use viable bacteria as a sustainable and concrete-embedded self healing agent was explored [14-16]. In one study spores of specific alkali-resistant bacteria related to the genus *Bacillus* were added to the concrete mixture as self-healing agent [16]. These spores germinated after activation by crack ingress water and produced copious amounts of crack-filling calcium carbonate-based minerals through conversion of precursor organic compounds which were also purposely added to the concrete mixture. However, in that study it was found that the bacteria-based self-healing potential was limited to relatively young (7-days cured) concrete only, as viability and related activity of bacterial spores directly (unprotected) embedded in the concrete matrix was restricted to about two months. The present study builds further on results reported in latter research paper [16]. Here, bacterial spores and organic mineral precursor compounds are packed in porous expanded clay particles prior to addition to the concrete mixture. It is hypothesized that protection of bacterial spores within porous light weight aggregates extends their viability period and thus concrete self-healing functionality when embedded in the material matrix.

VIABLE BACTERIA AS SELF HEALING AGENT

The bacteria to be used as self healing agent in concrete should be fit for the job, i.e. they should be able to perform long-term effective crack sealing, preferably during the total construction life time. The principle mechanism of bacterial crack healing is that the bacteria themselves act

largely as a catalyst, and transform a precursor compound to a suitable filler material. The newly produced compounds such as calcium carbonate-based mineral precipitates should act as a type of bio-cement what effectively seals newly formed cracks. Thus for effective self healing, both bacteria and a bio-cement precursor compound should be integrated in the material matrix. However, the presence of the matrix-embedded bacteria and precursor compounds should not negatively affect other wanted concrete characteristics. Bacteria that can resist concrete matrix incorporation exist in nature, and these appear related to a specialized group of alkali-resistant spore-forming bacteria [16]. Interesting feature of these bacteria is that they are able to form spores, which are specialized spherical thick-walled cells somewhat homologous to plant seeds. 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 (Fig. 1).

However, when bacterial spores were directly added to the concrete mixture, their life-time appeared to be limited to one-two months [16]. The decrease in life-time of the bacterial spores from several decades when in dry state to only a few months when embedded in the concrete matrix may be due to continuing cement hydration resulting in matrix pore-diameter widths typically much smaller than the 1- μm sized bacterial spores [16]. Another concern is whether direct addition of organic bio-mineral precursor compounds to the concrete mixture will not result in unwanted loss of other concrete properties. In the preceding study it was indeed found that various organic bio-cement precursor compounds such as yeast extract, peptone and calcium acetate resulted in a dramatic decrease of compressive strength. The only exception appeared to be calcium lactate what actually resulted in a 10% increase in compressive strength compared to control specimens [16]. In order to substantially increase the lifetime

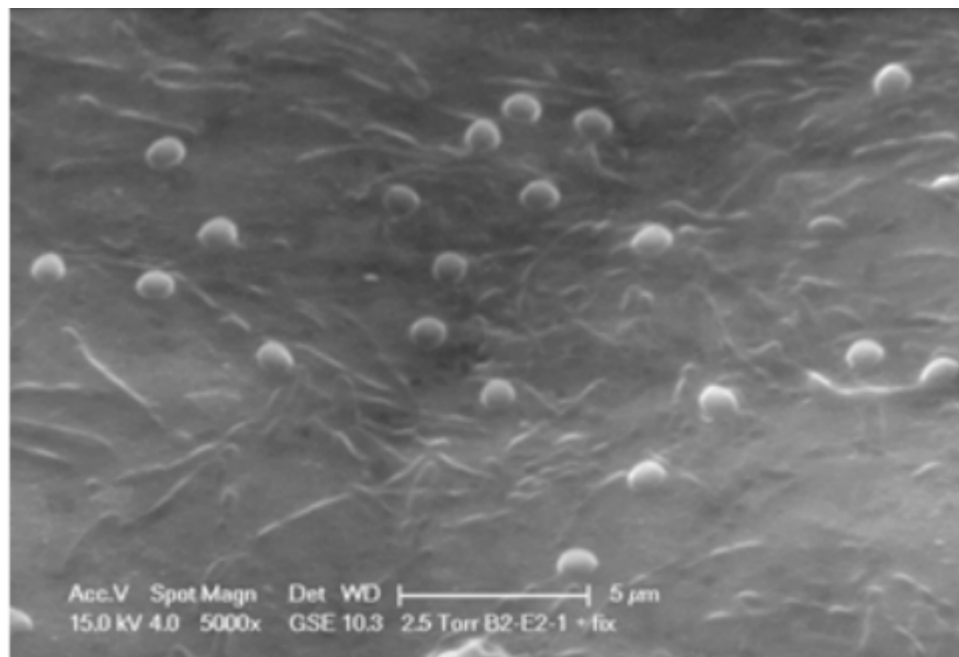


Figure 1. ESEM photomicrograph (5000x magnification) of alkali-resistant spore forming bacterium (*Bacillus* strain B2-E2-1). Visible are active vegetative bacteria (rods) and spores (spheres), showing that spore diameter sizes are in the order of one micrometer.

and associated functionality of concrete incorporated bacteria, the effect of bacterial spore and simultaneously needed organic biomineral precursor compound (calcium lactate) immobilization in porous expanded clay particles was tested in this study. It was found that protection of the bacterial spores by immobilization inside porous expanded clay particles before addition to the concrete mixture (Fig. 2) indeed substantially prolonged their life-time. Currently running viability experiments show that still after 6 months concrete incorporation no loss of viability is observed, suggesting that their long term viability as observed in dried state when not embedded in concrete is maintained. In subsequent experiments the expanded clay particles loaded with the two-component bio-chemical healing agent were applied as additive to the concrete mixture to test self-healing potential of bacterial concrete.

AUTONOMOUS CRACK REPAIR OF BACTERIAL SELF HEALING CONCRETE

Concrete test specimens were prepared in which part of the aggregate material,

i.e. the 2-4mm size class, was replaced by similarly sized expanded clay particles loaded with the biochemical self-healing agent (bacterial spores $1.7 \times 10^5 \text{ g}^{-1}$ expanded clay particles, corresponding to 5×10^7 spores dm^{-3} concrete, plus 5% w/w fraction calcium lactate, corresponding to 15 g dm^{-3} concrete). Before application, loaded expanded clay particles were oven-dried until no further weight loss due to water evaporation was observed (one week at 40°C). Control specimens had a similar aggregate composition but these expanded clay particles were not loaded with the bio-chemical agent. Both types of expanded clay particles (empty for control specimens and loaded for bacterial specimens) were

Composition of concrete specimens is shown in Table 1. The amount of light weight aggregate applied in this case represents 50% of the total aggregate volume. Replacement of such a high fraction of sand and gravel for expanded clay has consequences for strength characteristics of the derived concrete. In this specific case a 50% decrease in compressive strength was observed after 28 days curing when compared to specimens of similar aggregate

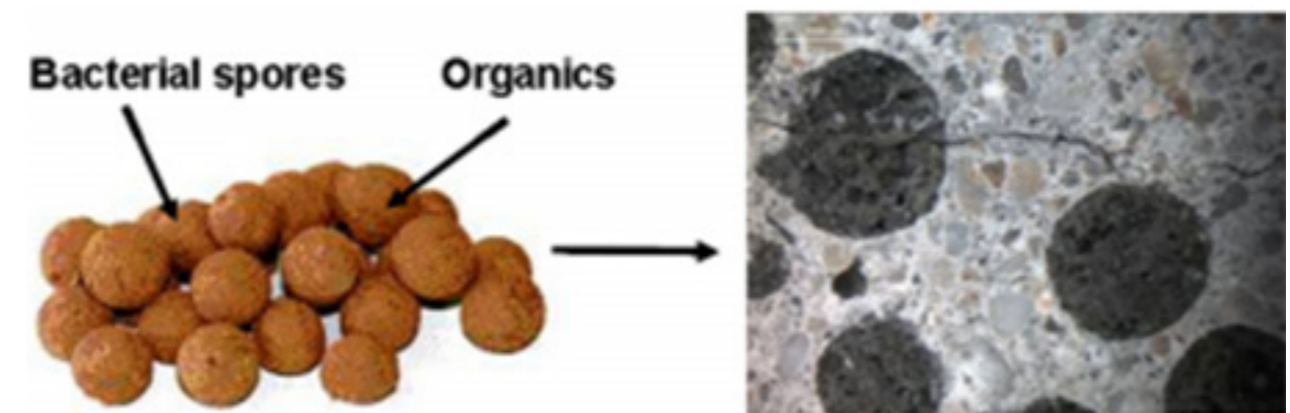


Figure 2. Self healing admixture composed of expanded clay particles (left) loaded with bacterial spores and organic bio-mineral precursor compound (calcium lactate). When embedded in the concrete matrix (right) the loaded expanded clay particles represent internal reservoirs containing the two-component healing agent consisting of bacterial spores and a suitable bio-mineral precursor compound.

Compounds	Volume (cm ³)	Weight (g)
2 - 4 mm LWA	196	167
1 - 2 mm LWA	147	125
0.5 - 1 mm Sand	147	397
0.25 - 0.5 mm Sand	128	346
0.125 - 0.25 mm Sand	69	186
Cement CEMI 42.5N	122	384
Water	192	192
Total	1001	1796

Table 1. Composition of concrete specimens. LWA refers to Liapor Sand R 1/4 expanded clay particles

composition without replacement of sand and gravel fractions for expanded clay particles. Although the expanded clay-based specimens featured a substantial decrease in strength, crack-healing capacity of specimens in which expanded clay particles were loaded with bacteria and organic mineral precursor compound (calcium lactate) appeared substantially improved. The self-healing capacity of pre-cracked concrete slabs sawed from 56 days (2 months) water cured concrete cylinders was determined by taking light microscopic images before and after permeability quantification. For the latter, pre-cracked concrete slabs were glued in an aluminum ring and mounted in a custom made permeability setup. Crack formation in concrete specimen slabs (10 cm diameter, 1.5 cm thickness) was achieved by controlled application of compressive-tensile stress (Fig. 3, left picture) at the 2 months cured specimens. Induced cracks featured crack length of 8 cm running from top to bottom of specimen and a crack width of 0.15 mm running completely through the 1.5 cm thick specimen. After crack induction, both sets (6 of each) of control (added expanded clay particles neither loaded with bacterial spores nor with organic compounds)

and bacterial concrete specimens (added expanded clay particles loaded with both bacterial spores and organic compounds) were submerged for two weeks in tap water at room temperature. Subsequently, permeability of all cracked specimens was quantified by automated recording of tap water percolation in time during a 24 hours period (Fig. 3). Comparison between bacterial and control specimens revealed a significant difference in permeability and thus in self-healing capacity. While cracks of all six bacterial specimens were completely sealed resulting in no measurable permeability (percolation of 0 ml water / h), only 2 out of six control specimens appeared perfectly healed. The four other control specimens featured permeability (water percolation) values between 0 and 2 ml/h. Microscopic examination of cracks (at the side of the slab being exposed to the water column) revealed that in both control and bacterial specimens precipitation of calcium carbonate-based mineral precipitates occurred. However, while in control specimens precipitation largely occurred near the crack rim leaving major parts of the crack unhealed, efficient and complete healing of cracks occurred in bacterial spe-

cimen as here mineral precipitation occurred predominantly within the crack itself (Fig. 4).

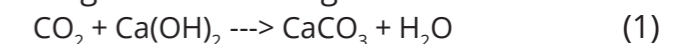
DISCUSSION AND CONCLUSION

The outcome of this study shows that crack healing of bacterial concrete based on expanded porous clay particles loaded with bacteria and calcium lactate, i.e. an organic bio-mineral precursor compound, is much more efficient than of concrete of the same composition however with empty expanded clay particles. The reason for this can be explained by the strictly chemical processes in the control and additional biological processes in the bacterial concrete. Non-hydrated cement particles exposed at the crack surface of concrete will undergo secondary hydration and in addition in control specimens carbon dioxide present in the bulk water will react with present portlandite (calcium hydroxide) particles to produce calcium carbonate-based mineral precipitates. Latter mineral precipitates will particularly form near the crack rim due to the relatively high solubility of calcium hydroxide. Here it is hypothesized that calcium hydroxide particles present at the surface of the crack interior will first scavenge all available carbon dioxide from crack ingress water, where after remaining calcium hydroxide will dissolve and diffuse out of the crack

into the bulk water. Once in the bulk water it will react with carbon dioxide present in close approximation to the crack rim resulting in the chemical production and precipitation of larger quantities of much lower soluble calcium carbonate.

Probable reason for the massive precipitation of calcium carbonate near the crack rim (Fig.4A) is that concentration of both reactants calcium hydroxide and carbon dioxide are relatively high here due to the opposing diffusion gradients of the respective reactants.

Calcium hydroxide diffuses away from the crack interior towards the overlying bulk water while carbon dioxide diffuses from the bulk water towards the crack interior where it is scavenged by high concentrations of calcium hydroxide. The process of chemical calcium carbonate reaction from dissolved calcium hydroxide occurs according to the following reaction:



The amount of calcium carbonate production inside the crack in control concrete specimens is likely only minor due to the low amount of CO₂ present in the limited amount of water present in the crack interior. The self healing process in bacterial concrete is much more efficient due to the active metabolic conversion of calcium lactate by the present bacteria:

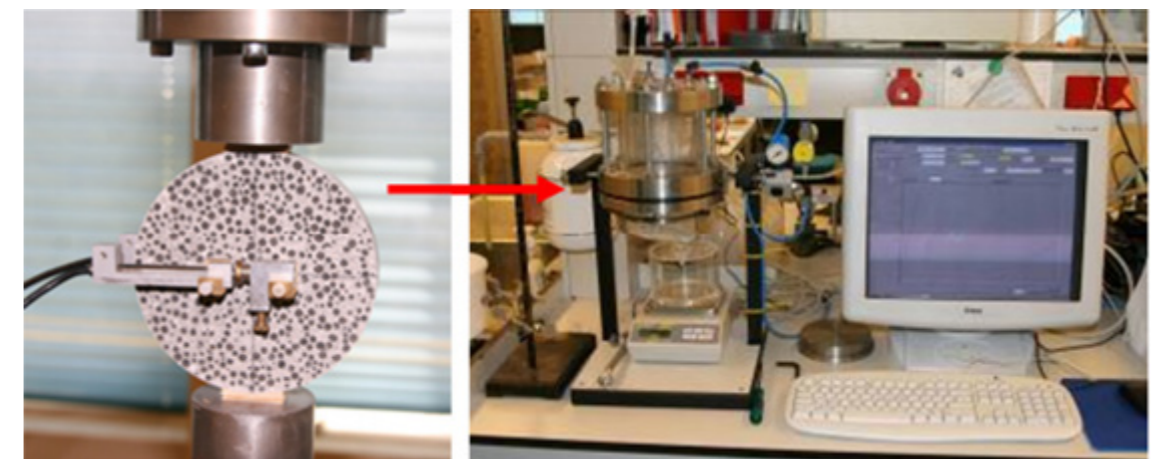


Figure 3. Pre-cracking of concrete slab and subsequent permeability testing

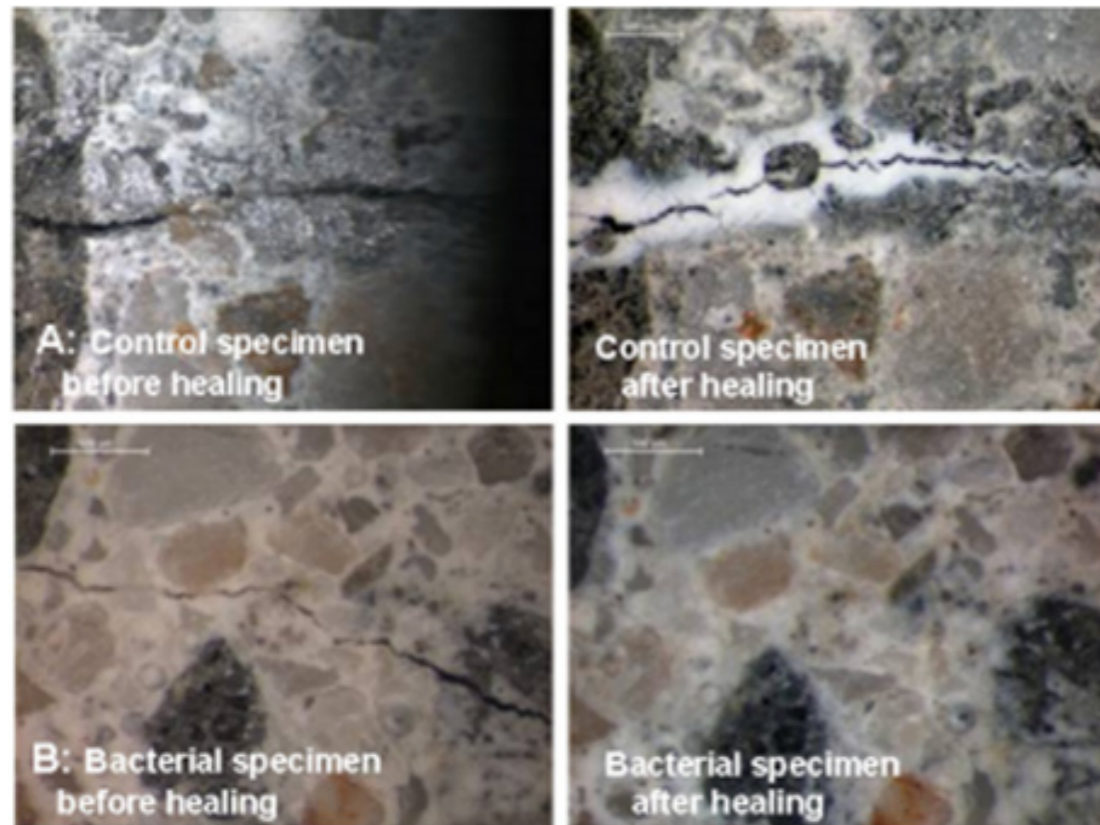


Figure 4. Light microscopic images (40 times magnification) of pre-cracked control (A) and bacterial (B) concrete specimen before (left) and after (right) healing (2 weeks submersion in water). Mineral precipitation occurred predominantly near the crack rim in control but inside the crack in bacterial specimens. Efficient crack healing occurred in all six bacterial and two out of six control specimens.

This process results in the precipitation of substantially higher amounts of calcium carbonate inside the crack as calcium carbonate is in this case not only directly produced from the conversion of calcium lactate in equimolar amounts of calcium carbonate, but also indirectly via the chemical reaction of metabolically produced CO₂. As latter carbon dioxide is produced at the surface of the crack interior it will directly react with portlandite particles still present in the crack interior. In the latter case, portlandite does not dissolve and diffuse away from the crack surface, but instead reacts directly on the spot with local bacterially produced CO₂ to additional calcium carbonate. The process of bacterial calcium lactate conversion thus results in the production of in total six calcium car-

bonate equivalents, resulting in efficient crack sealing as can be seen in Figure 4B. In this study the potential effect of only calcium lactate addition (without addition of bacterial spores) on crack healing was not considered. In order to establish the purely chemical effect of calcium lactate additions on crack-healing potential, experiments under completely sterile conditions have to be performed. This, however, is technically difficult considering the introduced effects of needed heat sterilization (120°C for 20 minutes) or chemicals on specimen characteristics. As self-healing tests in this study were performed under non-sterile conditions (realistic conditions) it can not be excluded that bacteria present in tap water used for curing, self-healing and permeability experiments contain

bacteria that may have (in addition to purposely added bacterial spores) contributed to metabolic conversion of calcium lactate to calcium carbonate-based minerals. One clear indication for metabolic (bacterial) conversion of calcium lactate, however, has been recently obtained in our laboratory by oxygen consumption measurements of concrete specimens. While bacterial spores and calcium lactate-containing specimens consumed substantial amounts of oxygen rapidly after submersion in water, strongly delayed oxygen consumption was observed in only calcium lactate-containing specimens, and no oxygen consumption occurred in only bacterial spores-containing specimens. As latter experimental data still need further quantification, it does suggest that addition of bacterial spores as part of the two-component biochemical healing agent may in fact not be necessary in cases where crack ingress water already contains bacteria able to metabolically convert calcium lactate. However, in order to assure rapid onset of crack-healing action and particularly to ensure their presence, co-application of calcium lactate and bacteria able to metabolically convert this compound appears the most favourable option.

Main objective of this study was to establish whether bacteria immobilized in porous expanded clay particles prior to concrete mix addition can substantially increase bacterially-mediated self-healing in comparison to direct unprotected addition of bacteria to the concrete mixture as was done in a previous study [16]. The results of this study appear promising as 100% healing (6 out of 6 tested specimens) of cracks induced in 2 months cured bacterial specimens occurred in contrast to 33% healing (2 out of 6 tested specimens) of control specimens. Tests showed furthermore that bacterial spore viability increased from 2 to more than 6 months when added immobilized (protected) inside porous expanded clay particles compared to direct (unprotected) addition to the con-

crete mixture. Ongoing experiments concern further quantification crack-healing, i.e. establishing the relationship between amount of healing agent added and effective healing of crack depth and width. From this study it can be concluded that active bacterially-mediated mineral precipitation can thus result in efficient crack-plugging and concomitant decrease in material permeability. The overall conclusion of this work is that the proposed two component bio-chemical healing agent, composed of bacterial spores and a suitable organic bio-cement precursor compound, using porous expanded clay particles as a reservoir is a promising bio-based and thus sustainable alternative to strictly chemical or cement-based healing agents, particularly in situations where concrete parts of a construction are not accessible for manual inspection or repair. However, before practical application becomes feasible, further optimization of the proposed system is needed. E.g., the amount of healing agent needed should be minimized in order to become economically competitive with currently existing repair techniques and furthermore to reduce consequences such as loss in compressive strength.

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