

Recent advances on self healing of concrete

E. Schlangen, H. Jonkers, S. Qian & A. Garcia
Delft University of Technology, Microlab, Delft, Netherlands

ABSTRACT: In this paper an overview is given of new developments obtained in research on self healing of cracks in cement based materials and asphalt concrete. At Delft University various projects are running to study self healing mechanisms. The first project that is discussed is Bacterial Concrete, in which bacteria are mixed in concrete, that can precipitate calcite in a crack and with that make concrete structures water tight and enhance durability. In a second project hybrid fiber reinforced cementitious materials are studied that can mechanically repair cracks when they occur. The last project described in this paper is on the raveling of porous asphalt concrete and how to heal this damage by incorporating embedded microcapsules or steel fibers. The state of the art results in all projects show that self healing is not just a miracle, but materials can be designed for it.

1 INTRODUCTION

In the past, quite some investigations on the topic of self healing of concrete have been conducted. Neville (2002) gives a useful overview of his literature search in this field. He puts the practical significance of autogenous healing in the reduction of water transport through cracks, for example in concrete water pipes. Neville also concludes from his literature research that there is no agreement between different studies about what happens inside the crack when self healing occurs and therefore further research would be useful. The early research on self healing of concrete mainly focused on water retaining structures or reservoirs where leakage through cracks was the main issue (Edvardsen 1999, Reinhardt & Joos 2003). In the research of Ter Heide et al (2005) and Granger et al (2006) the main focus was regaining mechanical properties of cracks in early age concrete by ongoing hydration of cement particles. Ter Heide (2005) gives a nice overview of different causes of autogenic healing (Fig. 1), in which a materials has already by nature the ability to heal itself.

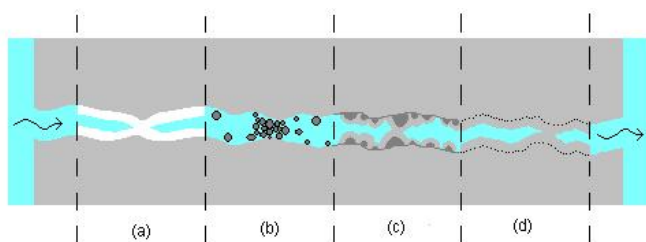


Figure 1. Possible causes of self healing: (a) formation of calcium carbonate or calcium hydroxide, (b) sedimentation of particles, (c) continued hydration, (d) swelling of the cement-matrix, (Ter Heide, 2005).

On the other hand, materials can also be designed to have a self healing capacity (Schlangen & Joseph 2008). Then we classify them as autonomic materials, which can again be subdivided in passive and active modes. A passive mode smart material has the ability to react to an external stimulus without the need for human intervention, whereas an active mode smart material or structure requires intervention in order to complete the healing process.

Van Breugel (2007) discusses the performance of structures with elapse of time. Gradual degradation occurs until the moment that first repair is urgently needed. The durability of concrete repairs is often a point of concern. Very often a second repair is necessary only ten to fifteen years later. Spending more money initially in order to ensure a higher quality often pays off. The maintenance-free period will be longer and the first major repair work can often be postponed for many years. The extreme case would be that no costs for maintenance and repair have to be considered at all because the material is able to repair itself.

In the last 5 years the design of materials with healing ability is becoming more and more popular in a wide range of materials and applications (Van der Zwaag 2007 and Gosh 2008). For cement based materials different method can be found in literature. In the first type of approaches encapsulated sealants or adhesives are used (Dry 2000). The adhesives can be stored in short fibre (Li et al. 1998, Qian et al. 2009) or in longer tubes (Nishiwaki et al 2006, Joseph 2008, Joseph et al. 2008). Another approach is incorporating an expansive component in the concrete which starts to expand and fill voids and cracks when triggered by carbonation or moisture ingress

(Hosoda et al. 2007, Sisomphon et al. 2009). Using bacteria to stimulate the self healing mechanism is an alternative but promising technique studied at different groups (Bang et al. 2001, Jonkers & Schlangen 2007, De Muynk 2008).

In this paper some recent advances in three ongoing projects at Delft University are discussed. First the Bacterial Concrete is described in which the main focus is sealing of cracks and thus blocking of the path to the reinforcement in order to improve the durability. Second topic is on promoting the healing of damage in fibre reinforced materials by adding different components to the mixture. The third project is on healing of asphalt concrete, where two smart methods of healing are studied. In one case porous particles containing a rejuvenator are used to start the passive autonomic healing process. In the other case steel wool fibres are mixed through the bitumen which can be heated with an induction machine. This internal heating can repair damage in the material. This is then an active autonomic healing method, because an external stimulus is needed to start the process.

2 SELF HEALING PROJECTS

2.1 *Bio-concrete*

In this study the potential of bacteria to act as a self-healing agent in concrete is investigated. Although the idea to use bacteria and integrate them in the concrete matrix may seem odd at first, it is not from a microbiological viewpoint. Bacteria naturally occur virtually everywhere on earth, not only on its surface but also deep within, e.g. in sediment and rock at a depth of more than 1 km. Various species of so-called extremophilic bacteria, i.e. bacteria that love the extreme, are found in highly desiccated environments such as deserts, but also inside rocks and even in ultra-basic environments which can be considered homologous to the internal concrete environment. Typical for many desiccation- and/or alkali-resistant bacterial species is their ability to form endospores. These specialized cells which are characterized by an extremely low metabolic activity, are known to be able to resist high mechanically- and chemically induced stresses and are viable for periods of up to 200 years. In some previously published studies the application of bacteria for cleaning of concrete surfaces (De Muynck et al. 2008) and strength improvement of cement-sand mortar (Bang et al. 2001) was reported. Although promising results were reported, the major drawback of the latter studies was that the bacteria and compounds needed for mineral precipitation could only be applied externally on the surface of the structures after crack-formation had occurred. This methodological neces-

sity was mainly due to the limited life-time (hours to a few days) of the (urease-based) enzymatic activity and/or viability of the applied bacterial species. In the present study the application of alkali-resistant endospore-forming bacteria to enhance the self-healing capacity of concrete is investigated. Tensile- and compressive strength characteristics of reference (no bacteria added) and bacterial concrete are quantified (Jonkers & Schlangen 2009a). Furthermore, the viability of bacteria immobilization in concrete is quantified and, finally, calcite precipitation potential of bacterial concrete is demonstrated by ESEM analysis. An example is given in Figure 2 where calcite crystals formed by bacterial precipitation are shown.

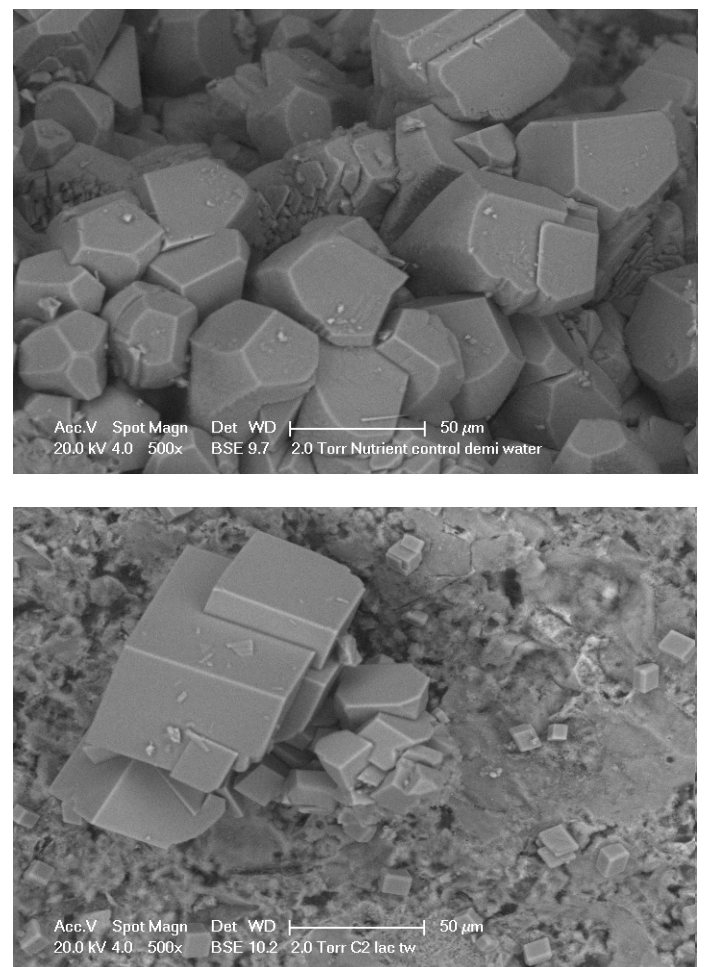


Figure 2. Biominerals observed by ESEM.

Extensive results of this study are published elsewhere (Jonkers et al. 2009b). To date the main conclusions of this ongoing research are that the experiments done in this study show that alkaliphilic endospore-forming bacteria integrated in the concrete matrix can actively precipitate calcium carbonate minerals. Water, needed for the activation of endospores, can enter the concrete structure through freshly formed cracks. Furthermore, for mineral precipitation, active cells need an organic substrate that can metabolically be converted to inorganic carbon that can subsequently precipitate with free calcium

to calcium carbonate. Free calcium is usually present in the concrete matrix, but organic carbon is not. In the first experiments organic carbon was applied externally as a part of the incubation medium, while ideally it should also be part of the concrete matrix. In that case only external water is needed to activate the concrete-immobilized bacteria which can then convert organic carbon present in the concrete matrix to calcium carbonate and by doing so seal freshly formed cracks.

Therefore it was decided to design a new strategy in which a two component biochemical healing agent composed of bacterial spores and a suitable organic bio-cement precursor compound is used. Both the spores and the food are immobilized in reservoir porous expanded clay particles. In this way the spores and bacteria are also protected during the production and hardening of the concrete and will survive longer until the moment that self healing is needed.

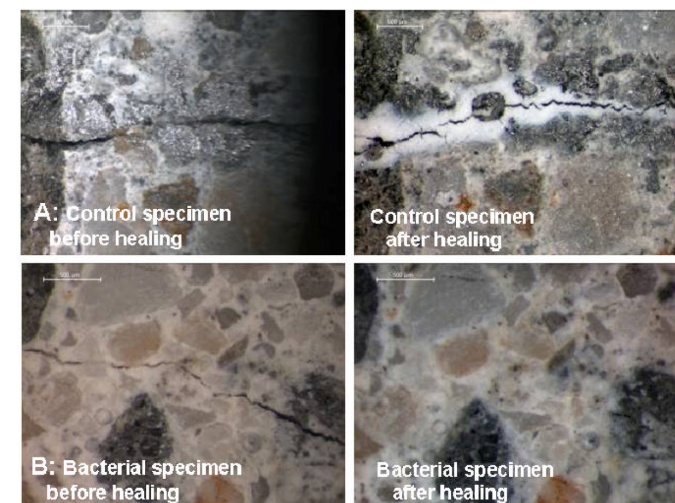
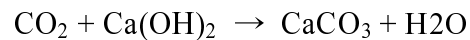


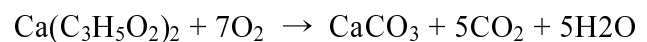
Figure 3. Cracked concrete specimens containing porous aggregates with food only (A) (top) and food plus bacteria (B) (bottom). The left figures are before and the right figures are after the healing.

Concrete disks are prepared containing the porous aggregates filled with food (lactate) only and with food and bacteria. The specimens are cured for 56 days and then tested in a deformation controlled tensile splitting loading to crack them partially. After this cracking the specimens are placed in a permeability test setup in which water is applied at one side of the specimen for 24 hours. After the healing the cracks are examined under the microscope and the results are shown in figure 3. Also the permeability of the healed specimens was determined. These results are discussed in Jonkers et al. (2009b). The outcome of this study shows that crack healing in bacterial concrete is much more efficient than in concrete of the same composition but without added biochemical healing agent. The reason for this can be explained by the strictly chemical processes in the control and additional biological processes in the bacterial concrete. On the crack surface of control

concrete some calcium carbonate will be formed due to the reaction of CO_2 present in the crack ingress water with Portlandite (calcium hydroxide) present in the concrete matrix according to the following reaction:



The amount of calcium carbonate production in this case is only minor due to the limited amount of CO_2 present. As Portlandite is a rather soluble mineral in fact most of it present on the crack surface will dissolve and diffuse out of the crack into the overlying water mass. Subsequently, as more CO_2 is present in the overlying water, dissolved Portlandite will as yet precipitate in the form of calcium carbonate but somewhat away from the crack itself, as can be seen in Figure 3A. The self healing process in bacterial concrete is much more efficient due to the active metabolic conversion of calcium lactate by the present bacteria:



This process does not only produce calcium carbonate directly but also indirectly via the reaction of on site produced CO_2 with Portlandite present on the crack surface. In the latter case, Portlandite does not dissolve and diffuse away from the crack surface, but instead reacts directly on the spot with locally produced CO_2 to additional calcium carbonate. This process results in efficient crack sealing as can be seen in Figure 3B.

The conclusion of this work is that the proposed two component biochemical healing agent composed of bacterial spores and a suitable organic bio-cement precursor compound, both immobilized in reservoir porous expanded clay particles, represents a promising bio-based and thus sustainable alternative to strictly chemical or cement based healing agents.

2.2 Self Healing fibre concrete

Besides permeability enhancement, many researchers also looked into the mechanical property recovery as a result of self-healing in concrete materials. As suggested by many previous studies (Edvardsen 1999, Ter Heide 2005, Reinhardt & Joos 2003), the crack width of the concrete material was found to be critical for self-healing to take place. The requirement of crack width to promote self-healing falls roughly below $200\text{ }\mu\text{m}$ and preferably lower than $50\text{ }\mu\text{m}$ (Edvardsen 1999), especially for self-healing based on ongoing hydration of cement. Yet in practice, such small crack width is very difficult to achieve consistently in normal concrete structures, if not possible at all.

To achieve controlled tight crack width, a new class of fiber reinforced strain hardening cementitious composites, termed as Engineered Cementitious Composites (ECC) has been developed by Li et al. (1998) and continuously evolved over the last 15 years. ECC has been deliberately engineered using micromechanics theory to possess self-controlled crack width that does not depend on steel reinforcement or structural dimensions. Instead, the fibers used in ECC are tailored to work with a mortar matrix in order to suppress localized brittle fracture in favor of distributed microcrack damage, even when the composite is tensioned to several percent strain. ECC with crack width as low as 30 micron have been made. Given the well controlled crack width, Li et al (1998) have investigated the self-healing behavior of ECC under a number of exposure conditions. In their experiments, deliberately pre-cracked ECC specimens were exposed to various commonly encountered environments, including water permeation and submersion, wetting and drying cycles, and chloride ponding. The mechanical and transport properties can be largely recovered, especially for ECC specimens preloaded to below 1% tensile strain. Besides the small crack width, the low water/binder ratio in addition to the large amount of fly ash in their mixture also helps promote self healing via continued hydration and pozzolanic activities.

Self-healing behavior of pre-cracked SHCC (strain hardening cementitious composites) is also a main research topic in the Microlab at Delft University. SHCC made with local waste materials (blast furnace slag and limestone powder) is investigated (Qian et al. 2009). Four-point bending tests are used to precrack SHCC beam specimens deflected up to 2.4 mm with subsequent curing in water and air for 28 days. The sample submerged in water shows greatly enhanced deflection capacity as well as stiffness recovery due to the healing products presented in the microcrack, while this is not the case for specimens cured in air. The ESEM and XEDS observation further confirmed the finding in mechanical tests. The mechanical properties are recovered after healing in an environment where water is present and the cracks are filled with reaction products, see Figure 4.

The following conclusions can be drawn based on this investigation (Qian et al. 2009):

1. For specimens submerged in water, the deflection capacity after self-healing can recover about 65% to 105% compared with those virgin specimens, while this ratio is about 40% to 60% for air cured specimens. Furthermore, the stiffness of initial linear stage of self-healed specimen is much larger compared with that of the air cured specimen due to the presence of healing products formed inside the crack and strengthened the bridging fiber.

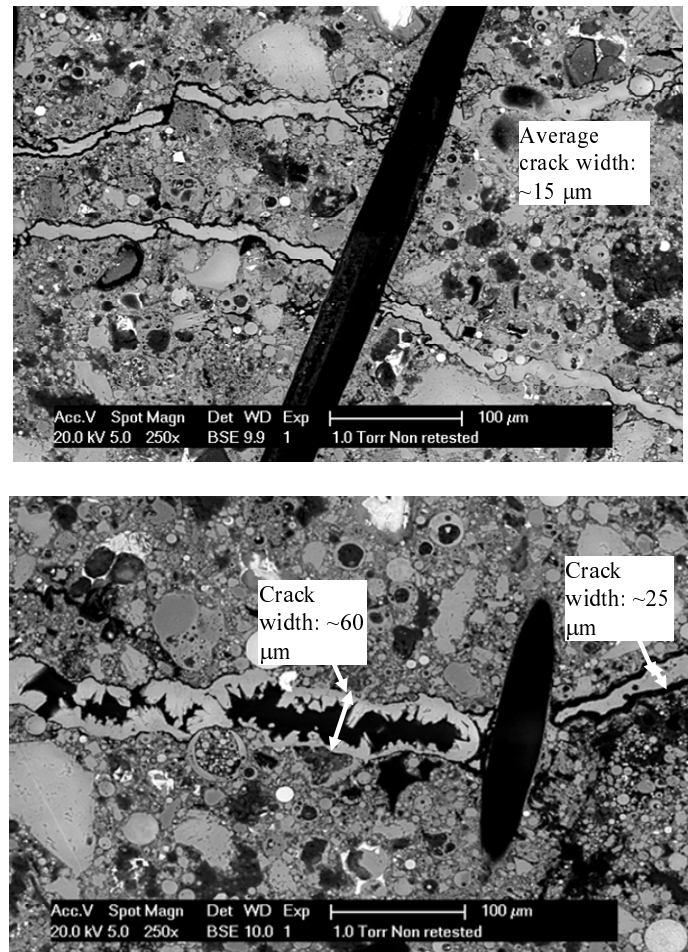


Figure 4. ESEM images of cracked SHCC samples cured in water.

2. The observations under ESEM and XEDS confirm that the microcracks submerged in water were healed mainly with calcium carbonate. ESEM also suggests that the healing products grow from both faces of the crack towards the middle of the crack. This may be explained by the relatively high concentration of calcium hydroxide near the crack surface via diffusion process from the bulk cementitious material and the fractal surface which may serve as the calcite nucleation sites.

3. Self-healing behavior in SHCC heavily depends on the availability of unhydrated cement and other supplementary cementitious materials. Low water/cementitious material ratio and high percentage of cementitious material appear to promote self-healing behavior.

4. Microcrack with smaller crack width as in the SHCC mixtures is preferable as far as continuous hydration-based self-healing is concerned, as it requires much less healing products to fill the crack and it is much easier for the healing products to grow from both faces of the crack to get connected.

Two main points that still need to be improved are the healing of larger cracks and the curing of specimens in air. In figure 4 it can be seen that small cracks with a width less than 15 μm are no problem to heal. However cracks around 60 μm in width can only partially be filled with healing products. To re-

duce the crack width further the effect of adding microfibers to the PVA-fibre reinforced cementitious composite is investigated (Antonopoulou 2009, Tziviloglou 2009). The microfibers used are steel wool or rockwool fibres with a length of 2 mm and an average diameter of 8 micron. These small fibres help to distribute the cracks in the cement matrix even more and thus the result is smaller cracks, which improves the self healing capacity.

The second negative point is that the self healing mechanism of ongoing hydration only works if water is present. To promote self healing also in a dry environment two approaches are under investigation. Hollow plant fibres, due to large storage volume for liquid, can potentially be used for a new self healing concrete system. The idea is to use the plant fibre as a reservoir for healing agent, which can be water to get ongoing hydration or which can also be a glue. Once a crack occurs, the healing agent will follow or diffuse toward the crack and eventually heal the crack. From the investigation, it was discovered that self healing of concrete cracks would be potentially feasible using coated wood fibres (or actually fibre bundles) that are filled with healing agent. The fibre bundles have a diameter of around 200µm and a length of about 10 mm (see also Sierra Beltran & Schlangen, 2010). For the proposed self-healing system to work, it is crucial that the fibres must be broken to deliver the healing agent. In this investigation, the wood fibre bundles were first coated with polysiloxane coating, then filled with a fluorescent dye solution and finally sealed properly. The treated fibre bundles were then fractured. The coated wood bundles tend to fail in a delamination mode (see figure 5) along their length and negative pressure force caused by the sealed ends is not an issue, as it might be with continuous or short fibres that fail in a brittle way with a single crack plane (Joseph 2008). As a result, healing agent could be released from the splintered fibre bundles into the damaged areas where it subsequently repairs.

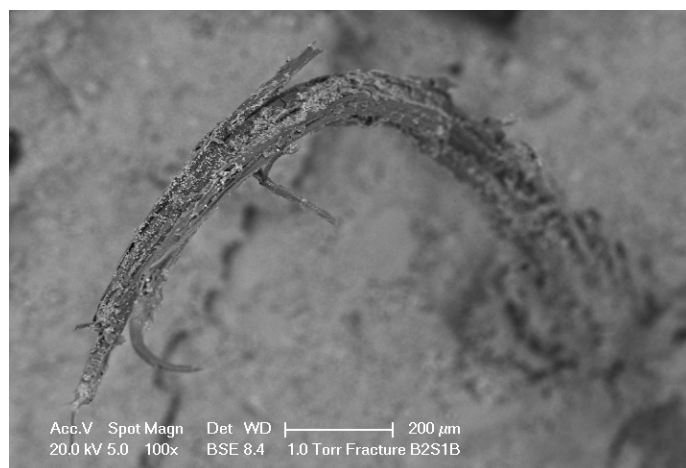


Figure 5. ESEM picture of a fibre-bundle broken in a delamination mode.

A second option to promote self healing in a dry environment which is under investigation is the use of Super Absorbent Polymers (SAP) in the mix. The SAP's are filled with water during the mixing process and form in such a way water pockets in the concrete that can be used for hydration of the cement and thus self healing in a later stage. SAP's are known as additive to mitigate autogenous shrinkage in concrete (Jensen & Hansen 2001). The self healing capacity of the SHCC is already improved by adding these SAP's in specimens that are cracked and subsequently stored in water (Antonopoulou 2009, Tziviloglou 2009). However these SAP's can also work for specimens stored in air. The water-pockets probably are emptied during or shortly after the first hydration. When the material cracks at a later stage no water is left anymore. But after some rain on the structure the SAP's located in the cracked zone are again filled and then slowly release the water for the self healing mechanism. This seems to be a realistic and practical scenario which is currently under investigation.

To optimize the self healing mechanisms in SHCC materials a model is developed (Schlangen et al. 2009) that is uses discrete fibers and can simulate distributed cracking and ductile behaviour as shown in figure 6.

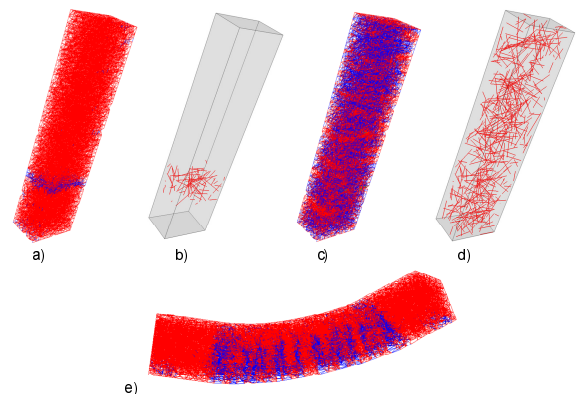


Figure 6. Simulations of SHCC material with fibres containing healing agent.

In figure 6 an example of the simulations of a tensile test and a four-point bending tests are shown. In the model of figure 6a and 6b only fibres with healing agent are used. Figure 6a shows the crack pattern and figure 6b shows the fibres from which healing agent is released. In figure 6c and 6d a hybrid material containing mechanical fibres to reduce crack width and distribute cracks as well as healing fibres containing a healing agent are used. It is shown that in the case distributed cracking is obtained much more fibres in the material are activated and the healing liquid is used in a much more efficient way. Figure 6e shows a bending test of a specimen containing fibres with mechanical fibres and healing agent. The simulations are used to investigate the amount of fibres and the amount of healing agent that is optimal.

2.3 Healing Porous Asphalt Concrete

After some years, asphalt binder is degraded by environmental factors, especially due to UV-radiation from the sun, until it loses the ability to bind the surface particles together. This results in cracks which allow damaging moisture into the lower pavement levels, creating surface roughness, pot holes, degradation and eventual structural failure. At present, there are no solutions to close cracks in the pavement. Occasionally, when signs of ageing are visible, a sealant that protects asphalt surfaces from environmental degradation and moisture penetration is applied to the surface. Other times, asphalt rejuvenators, with the capability of changing the chemical composition of bitumen, are applied to the surface. All these procedures can increase the lifetime of asphalt for several years before rehabilitation or reconstruction is required, but they have the disadvantage that they only work in the first centimeters from the surface and can reduce sliding resistance.

It is generally known that asphalt roads can heal by themselves, but it is a slow process at ambient temperature, and it only works if there is no traffic circulation on the road. It is also well known that the amount of healing increases when the material is subjected to a higher temperature during the rest period (Bonnaure et al. 1982). Therefore a project was started to investigate the increase of the self-healing rates of the road, which is as a good method to increase the lifetime of the pavement. As a simplification, bitumen could be considered as a very dense oil; when a crack appears in it, it will close by itself, but it will do much faster if the “liquid behavior” of bitumen is increased. That can be done by increasing its temperature or by mixing with a less dense oil.

In this paper, two fairly new ideas are presented: Induction heating of asphalt concrete and microcapsules filled with a healing agent (Garcia et al. 2009a). Both of them to increase the self-healing rates of asphalt concrete and hereby, the lifetime of the road.

2.3.1 Capsule-method

Bitumen can be considered as a two phase material with a liquid phase, called maltenes, and a solid phase, called asphaltenes. With time, the liquid phase is oxidized, disappearing and causing asphalt to become dry and brittle. To avoid this, maltenes have been traditionally applied on the road surface once signs of ageing start appearing. The problem is that this type of treatment is superficial, with what only the first centimeters from the surface are affected. To solve this, it was thought that the optimum way of adding maltenes to the road would be by mixing capsules filled with maltenes with the asphalt concrete. With this, aging effects could be avoided over the complete depth of the pavement. In Figure 7 a

scheme is shown with the effect of the capsules on the asphalt concrete. When a crack close to a capsule occurs in the material of the road, the capsule will break and the maltenes will be in contact with the bitumen around. Then, by diffusion both, maltenes and damaged bitumen will be mixed. The bitumen will be rejuvenated and the crack will be easily closed. With this autonomic method of using capsules the self healing rate is increased a lot compared to the autogenic self healing capacity of the asphalt.

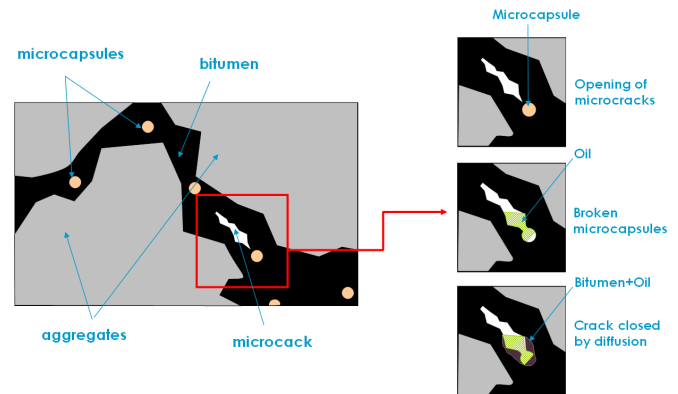


Figure 7. Schematic representation showing the working of encapsulated oil to heal cracks in asphalt concrete.

Many researches explain how to make capsules, but asphalt concrete is a real harsh environment for the capsules to survive. These capsules should encapsulate very viscous hydrocarbons based oil, they should not react with bitumen, and they should resist the mixing process with the aggregates and the bitumen and the compaction on the road at about 180°C. Besides, they should not be so resistant that they never break. To solve this, maltenes have been encapsulated in very porous sand and covered with a composite made of a thermoresistant resin and very fine sand. Research to prove the self healing capacity of this new material incorporating these capsules is ongoing.

2.3.2 Induction Heating

The basis for the second approach that is followed is heating the asphalt with induction energy to increase its healing rate. The first prerequisite of induction heating is that the heated material must be conductive. In many previous studies it has been demonstrated how it is possible to make asphalt or concrete conductive by adding electrically conductive fillers and fibers (Garcia et al. 2009b). The second prerequisite is that these fillers and fibers are connected in closed-loop circuits. In Figure 8 a schematic representation is given of the system in which inductive energy is used for the healing of asphalt concrete. First a microcrack appears in the bitumen. If enough volume of conductive fibers or fillers is added they will form closed-loops circuits all around the microcrack. Then, if this magnetically susceptible and electrically conductive material is placed in the vi-

cinity of a coil, eddy currents are induced in the closed-loops circuits, with the same frequency of the magnetic field. Heat is generated through the energy lost when eddy currents meet with the resistance of the material and, finally, bitumen is melted and the crack is closed.

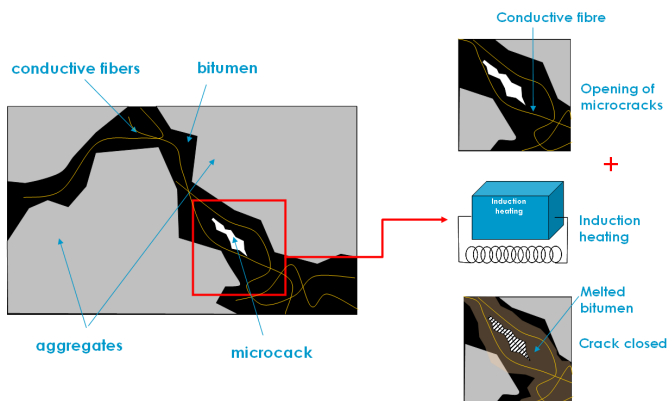


Figure 8. Schematic representation showing the system of using fibres heated with induction energy to heal cracks in asphalt concrete.

This research is being conducted in three steps. First, the optimum combination of conductive fibers and fillers was chosen; then, the temperature reached depending on the electrical conductivity was found. Finally to prove the healing, the samples will be repeatedly broken in three point bending and heated and healed again. It has been found that electrically conductive fibers are much more efficient than fillers when increasing the conductivity and that there is an optimum in the electrical conductivity, related to a certain volume of fibers, above which it is difficult to increase the conductivity. It has been discovered also that although it is not necessary that the sample is conductive to heat it, the optimum volume of fibers for heating coincides with the optimum volume of fibers to have the maximum conductivity.

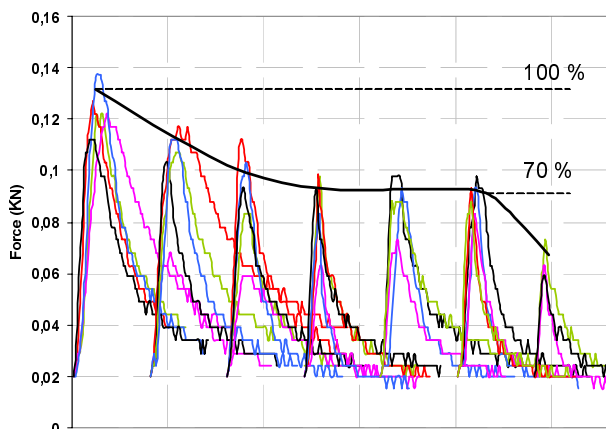


Figure 9. Force-deformation diagrams of bitumen specimen with steel-wool fibres loaded in 3 point bending, in 6 cycles of loading-heating and reloading.

Figure 9 gives an indication of the healing capacity of the bitumen. The figure shows the load-deformation curves of four samples and the stress-strain curves of

these samples after 6 heating, healing and reloading cycles. The samples were frozen at -20°C during the tests to avoid creep; so the test specimens had brittle fracture (the elastic modulus is clearly appreciated in the curves). During the healing process (induction heating) it could be seen that the cracks disappeared. In Figure 9, the resistance of the samples after the fifth healing is about 70 % of the original one. Besides, the elastic modulus is very similar, but the slope after the ultimate strength is steeper in the healed samples. This is logic because the healed zone is a weak zone where all the fibers are broken, so the sample is more brittle in that section.

3 DISCUSSION AND CONCLUSIONS

In this paper self healing techniques in three different materials are discussed. The first application is using Bacteria to precipitate calcite in cracks in concrete. With this method relatively large cracks in reinforced concrete can be filled. The method does not lead to strength improvements of the structure, but by filling the crack, the path to the reinforcement is blocked. Herewith the ingress of liquids and ions that start reinforcement corrosion is stopped and thus the durability of the structure is enhanced. Furthermore this method is useful for water retaining structures. Cracks can be filled in this way and leakage can be stopped. Especially in underground structures where repair is difficult or impossible Bacterial concrete has a big future.

In the second application SHCC materials are studied, which have already a high potential for self healing because of their small crack widths. New additions, like microfibers and SAP's even promote this self healing capacity further.

The third application is for asphalt concrete in which the self healing capacity is enlarged by using encapsulated oil and micro-steel fibres. The latter approach has been proven to work in the laboratory and will be applied in a real road in the Netherlands in 2010.

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