Use of encapsulated healing agents to limit water uptake through cracks in mortar

VAN BELLEGHEM Bjorn[1,2,a], VAN TITTELBOOM Kim[1,b], DE BELIE Nele[1,c *]

[1]Magnel Laboratory for Concrete Research, Department of Structural Engineering, Faculty of Engineering and Architecture, Ghent University, Technologiepark 904, BE-9052 Zwijnaarde, BELGIUM
[2]Strategic Initiative Materials (SIM vzw), project ISHECO within the program 'SHE', SIM vzw, Technologiepark 935, BE-9052 Zwijnaarde, BELGIUM

[a]Bjorn.VanBelleghem@UGent.be, [b]Kim.VanTittelboom@UGent.be, [c]Nele.DeBelie@UGent.be

*corresponding author

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Abstract. Crack formation is a common issue that leads to durability problems in concrete structures. Fluids containing aggressive substances can rapidly penetrate in the concrete matrix through the cracks and cause steel corrosion or deterioration of the concrete. In order to avoid major damage to the concrete structures, repair of cracks is in most cases necessary. Creating a self-healing cementitious material is a possible solution to avoid major repair works of concrete structures. In this research, autonomous healing of cracks by encapsulated healing agents was investigated to reduce water ingress through cracks in mortar.

The two polymeric healing agents that were used in this study were able to reduce the water ingress through cracks. The low viscosity polyurethane created a complete and consistent crack healing, reducing the water absorption to values even lower than uncracked cementitious material. For the high viscosity polyurethane the results showed more scatter due to uncomplete crack healing for some specimens. The reduction of water ingress due to the incorporation of the self-healing mechanism has a positive effect on the durability of cementitious materials and hence can prolong the service life of concrete structures.

Introduction

One of the major causes of durability issues in reinforced concrete structures is the appearance of cracks in the concrete matrix. Cracks will mainly occur at locations where the concrete is subjected to tensile stresses, which is almost always the zone where the steel reinforcement is placed. Due to cracking, there are preferential pathways for moisture to rapidly penetrate into the concrete onto the level of the steel reinforcement. Moreover, water will commonly act as a medium for the transport of other aggressive substances (e.g. chloride ions in marine environments) which can initiate corrosion of the reinforcing steel. Real concrete structures are almost never fully saturated. Therefore, the dominant ingress mechanism for water into concrete is capillary absorption [1], especially when cracks are present [2, 3]. Hence, capillary water absorption of cementitious materials is the main topic of investigation in the present study.

Repair works of cracks in concrete structures cause large costs, but besides that, parts of the structure where the cracks appear may not be accessible. Therefore, the idea of giving the concrete matrix the property of restoring the cracks itself, so called self-healing concrete, grew during the last two decades [4, 5]. At Ghent University the development of self-healing concrete has become a growing research topic during the past eight years. Several methods have already been investigated and one of the promising approaches is autonomous crack healing by means of encapsulated healing agents [6, 7]. This approach requires a set of discrete brittle capsules filled with a healing agent which are placed in the cementitious matrix. Upon the appearance of a crack, the brittle capsules break and the liquid healing agent is released in the crack. The used healing agents are usually polymers which contain isocyanate groups. When the healing agent is released in the crack, these isocyanate groups react with the moisture in the cementitious matrix to cause an exothermic polymerization reaction [8]. In this way the healing agent solidifies in the crack.

In this research, capillary water absorption tests were performed on mortar specimens with and without self-healing properties. In this way the efficiency of the self-healing approach is evaluated regarding the ingress of water. In previous research [9], self-healing of artificial cracks, created by thin plates, was investigated and it appeared that the technique is very promising for reducing the water absorption through cracks. The present study involves the evaluation of the autonomous healing of realistic cracks, mechanically created in the specimens.

Materials

Mortar. Ordinary Portland cement mortar with a water-to-cement ratio of 0.5 was used for all specimens in this research. The composition of the mortar was based on the standard mix as described in EN 196-1 [10].

Capsules. Encapsulation of the healing agents for the self-healing mortar was done by cylindrical borosilicate glass capsules with a length of 50 mm, an internal diameter of 3 mm and an external diameter of 3.35 mm. In practice, the use of glass as an encapsulation material is only feasible when the capsules are placed in the mold of a prefabricated element. However, further research is being conducted to select other encapsulation materials so that the capsules are able to survive the mixing process of the concrete.

Healing agents. Two different polyurethanes were used as healing agent in this study. The first healing agent is a non-commercial polyurethane developed in the framework of the SHEcon project and has been evaluated before as healing agent for artificial cracks [9, 11]. This polyurethane has a high viscosity of 6700 mPas at 25°C. It will be named PU_HV (High Viscosity) in this paper. The second healing agent is a commercially available polyurethane with a very low viscosity of 200 mPas at 25°C. This polyurethane will be named PU_LV (Low Viscosity) and has also been evaluated before for its efficiency in self-healing cementitious materials [7, 11, 12]. Both healing agents are one-component polyurethanes that react upon contact with moisture in the cementitious matrix.

Mortar specimens with(out) self-healing properties. Mortar prisms with dimensions of 160 mm x 40 mm x 40 mm were cast in wooden formworks. All specimens contained two reinforcement bars with a diameter of 3 mm. The bars were positioned at 10 mm from the bottom of the specimens and at 10 mm from the sides, creating a 20 mm spacing between the bars. In case of specimens with self-healing properties, two capsules filled with one of the polyurethanes were glued on thin threads between the reinforcement bars in the center of the prism (Fig. 1). After preparation of the molds, mortar was prepared and brought into the molds in two layers of 20 mm which were compacted separately on a vibrating table. Subsequently, the specimens were put in an air-conditioned room with a temperature of 20 ± 2°C and a relative humidity of more than 95% for 24 hours. After that period, the specimens were demolded and placed back in that room for 28 days (curing period). A total of 12 specimens was prepared: 3 sound mortar prisms without a crack (UNCR), 3 mortar prisms without self-healing properties where a crack was created (CR), 3 mortar prisms with embedded capsules filled with the high viscosity polyurethane (PU_HV) and 3 mortar prisms with embedded capsules filled with the polyurethane with low viscosity (PU_LV).

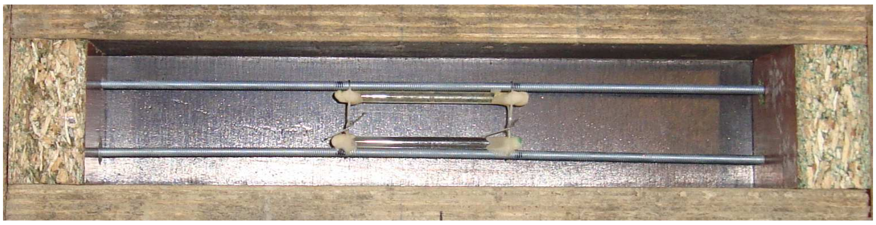

Figure 1: Mold preparation for a mortar prism with self-healing properties. Two polyurethane filled capsules glued on nylon threads in the center of the prisms.

Methods

Crack creation and healing. Cracks were created in the mortar prisms by means of three-point bending. The crack width was registered during the bending test by a linear variable differential transformer (LVDT). A target crack width of 100 µm was set for all specimens. Therefore, the specimens were loaded until a crack width of 400 µm was obtained. This final value was chosen based on previous experience. After that, the specimens were unloaded and due to the presence of the reinforcement bars the crack width decreased again to a value of around 100 µm. The final crack width value was then measured microscopically. For the specimens containing capsules filled with polyurethane, crack creation caused the capsules to break. Breakage of the capsules could be noticed during the bending test by popping sounds. Moreover, the moment of capsules breakage was always visible in the force-displacement curve of the bending test by a sudden small drop in the force.

Crack width measurements. As mentioned in the previous section, the crack width reduced after unloading the specimens. The crack widths were therefore measured microscopically using a stereomicroscope. The crack width was measured seven times along the length of the crack with an intermediate distance of 5 mm. Afterwards, the mean value of these seven measurements was calculated.

Specimen preparation. After crack creation, all specimens were dried in an oven at 40 ± 2°C. When the specimens had reached a constant mass, defined as a mass change less than 0.1% in 24 hours, they were taken out of the oven and placed in a climate room at a temperature of 20 ± 2°C and a relative humidity of 60% for a period of 24 hours prior to the capillary absorption test.

Once the specimens were taken out of the oven, the side surfaces were partly covered with aluminum butyl tape. Next, the test face of the specimens was covered with the same tape so that a region of 40 mm x 10 mm around the crack was exposed to water during the absorption test (Fig. 2). In that way only the water uptake through the crack was studied. In the case of uncracked reference samples, the tape was applied in the same way so that the same area of mortar was exposed to water.

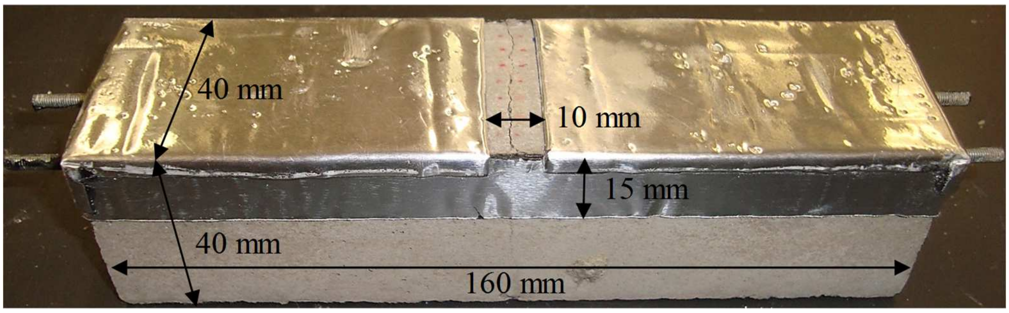

Figure 2: Cracked mortar specimens covered with aluminum butyl tape. The top surface in the figure is the test face of the specimen.

Capillary absorption test. A capillary water absorption test was performed in a climate room at 20 ± 2°C and 60% relative humidity based on the European standard NBN EN 13057 [13]. The specimens were placed one by one with their test face on line supports in a container containing water with an immersion depth of 2 ± 1 mm. At several time intervals the mass of the specimens was registered by weighing them on the scale with an accuracy of 0.01g. When the specimens were taken out of the container, they were first wiped with a wet cloth to remove surplus water before they were placed on the scale. After weighing, the specimens were returned to the tray. The mass of the specimens was recorded every half hour during the first eight hours of the test and once again after 24 hours.

Results and discussion

Autonomous crack healing. Almost immediately after breakage of the capsules, the polyurethane precursor was released in the crack, where it hardened due to the reaction with moisture. For the specimens where crack healing occurred with the low viscosity polyurethane (PU_LV), some leakage of polyurethane at the mortar surface already occurred during the three-point bending test. An example of the crack healing with this polyurethane is given in Fig. 3a. It can be seen that indeed a large part of the mortar surface next to the crack is covered with polyurethane. This can be attributed to the fact that there are high capillary forces that draw the polyurethane in the crack due to the limited crack width, but the crack volume is relatively small. Hence, combined with the low viscosity of the polyurethane, part of it will flow out of the crack. If the crack width would be larger, e.g. 300 µm, less leakage of polyurethane at the surface of the specimens would be noticed. In spite of this leakage however, it can be seen visually that the crack is well healed along the whole length.

In case the crack is healed with the high viscosity polyurethane (PU_HV) no real leakage of the polyurethane at the surface is found due to the high viscosity (Fig. 3b). Instead, a foaming reaction occurred during hardening. This foaming reaction can be seen at some places along the crack path. Due to this reaction and the high viscosity, the crack was not always completely healed along the whole crack length. Especially near the edges of the specimens, the crack sometimes remains unhealed (Fig. 3b). This will have an effect on the capillary water absorption through the cracks (see infra).

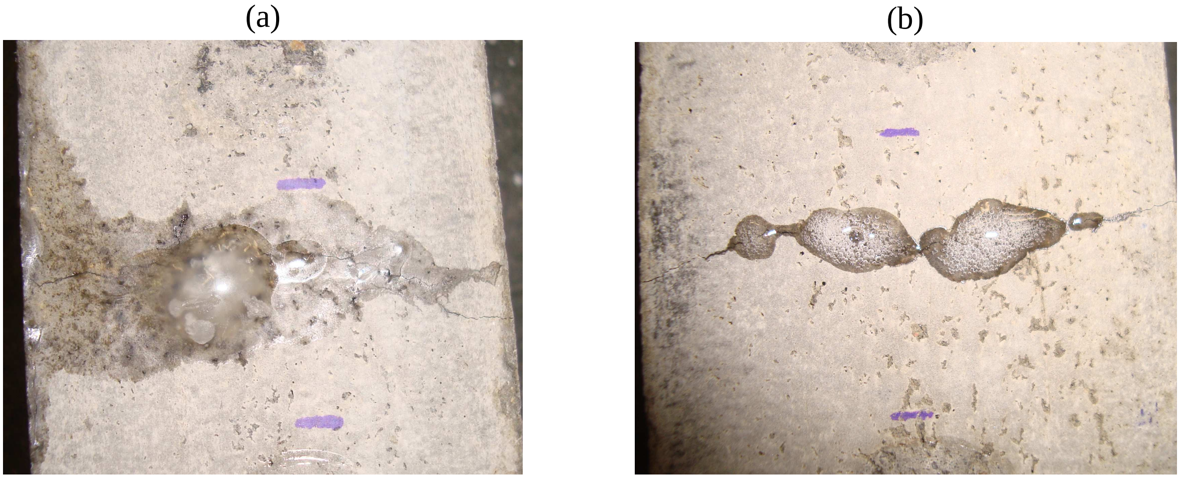

Figure 3: An example of (a) a specimen where a crack is healed by PU_LV and (b) a specimen where a crack is healed by PU_HV.

Capillary water absorption. The cumulative water absorption per unit surface area [g/cm²] was calculated and plotted in function of the square root of immersion time [$h^{0.5}$]. The mean cumulative water absorption curves of the three specimens in each series are given in Fig. 4.

(Un)cracked mortar. The huge influence of a relatively small crack on the ingress of water can be seen very clearly in Fig. 4. At the beginning of the absorption test the water enters very rapidly into the cracks. At the first measurement, after half an hour of exposure to water, the mean total water absorption of specimens with a crack is already more than seven times higher than for uncracked specimens. After the first half hour the water absorption rate in the cracked specimens decreases as the specimens get more and more saturated. At the end of the test, after 24 hours, the total water absorption is a little more than three times higher for specimens with a crack compared to the uncracked specimens. Clearly, there is a need for repair of cracks in order to prevent water and other aggressive substances to rapidly penetrate into the mortar matrix.

Mortar with autonomously healed cracks. Crack healing of relatively small cracks with PU_HV results in a capillary water absorption that is in between the water absorption of uncracked and cracked specimens (Fig. 4). The large standard error of this absorption curve is attributed to the fact that no complete crack healing was found these specimens. As mentioned before, this polyurethane showed a foaming reaction along parts of the crack, but near the edges of the specimen the crack sometimes remained unhealed. Crack healing with PU_LV resulted in a more complete crack healing. The cumulative absorption curve is located completely under the curve of uncracked mortar. The reason for this very high healing efficiency was the fact that a big part of the mortar surface, which was exposed to water during the absorption test, was covered with polyurethane (see Fig. 3a). After the first eight hours of the test however, it can be seen in Fig. 4 that the water absorption rate of the specimens where cracks were healed with PU_LV started to increase. A possible reason for this increase after the first eight hours of water absorption is that due to the low viscosity of the polyurethane, only the bottom part of the crack was healed. Once the water has penetrated next to the crack into the mortar matrix, it might reach the upper part of the crack that was not healed well. Consequently, water uptake increased again through this unhealed part of the crack. To investigate this hypothesis, further research is required to verify whether the upper part of the crack is indeed left unhealed. This can be done by splitting the specimens and evaluating the crack faces for polyurethane coverage [7].

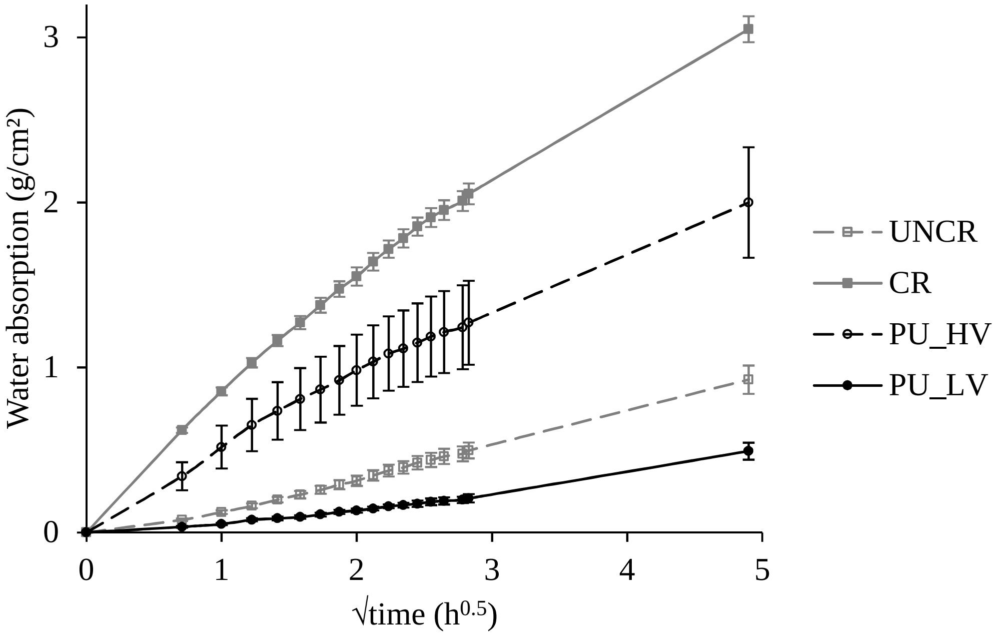

Figure 4: Cumulative water absorption curves in function of the square root of time for uncracked, cracked and healed specimens. Error bars represent the standard error on the mean values.

Sorption coefficient. The gradient of the line from the intercept of the water absorption curve to the cumulative mass of the water uptake per unit area recorded at 24 hours was defined as the sorption coefficient S [$kg/m^2/h^{0.5}$] according to the standard NBN EN 13057 [13]. In the next sections the sorption coefficients of the different specimen series will be compared as a method for evaluating the healing efficiency.

Fig. 5 shows the results of the capillary sorption coefficients of all individual specimens in function of their crack width. First, the results of the uncracked (crack width zero) and cracked mortar specimens will be discussed (white and grey squares respectively in Fig. 5). For uncracked mortar a mean sorption coefficient of 1.89 $kg/m^2/h^{0.5}$ was found. The presence of a relatively small crack (< 100 µm) has a big influence on the capillary water absorption. The sorption coefficient of the mortar prism with the smallest crack that was tested in this research (62 µm), was already more than three times higher than the sorption coefficient of uncracked mortar.

The sorption coefficients of the mortar prisms where the cracks were healed by the high viscosity polyurethane (PU_HV) or the low viscosity polyurethane (PU_LV) are also given in Fig. 5 in function of the crack width (white and black circles respectively). The capillary sorption of all healed specimens is clearly lower than for cracked mortar. In case of PU_HV there is a large scatter in the results. As mentioned before, this was due to an incomplete healing of the cracks. In case of crack healing with PU_LV the healing performance is much more similar for all specimens than for crack healing with PU_HV. The water absorption of specimens with healed cracks was even lower than the water absorption of uncracked mortar. As mentioned before, this is mainly due to the fact that some polyurethane hardened on the mortar surface and consequently covered part of the mortar surface during the absorption test. Nevertheless, it can be concluded that there was always a complete crack healing.

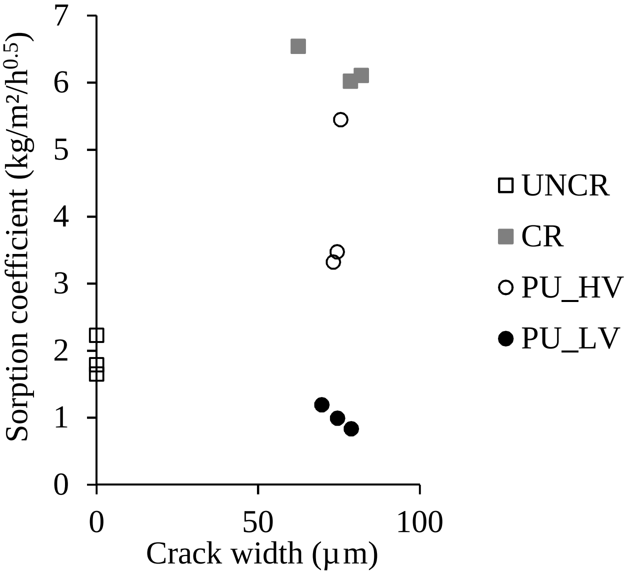

Figure 5: Capillary sorption coefficient in function of crack width for uncracked specimens, cracked specimens and specimens healed by PU_HV or PU_LV.

Self-healing efficiency. In order to make clear conclusions about the performance of the self-healing cementitious materials, the self-healing efficiency was determined [14]. The self-healing efficiency of a mortar specimen [%] was defined as the ratio of the difference between the mean sorption coefficient of the cracked specimens and the sorption coefficient of the healed specimen over the difference between the mean sorption coefficient of the cracked and the uncracked specimens.

When the mean values of the sorption coefficient of the cracked and uncracked specimens were determined, the self-healing efficiency of the autonomously healed specimens could be calculated. For two specimens where crack healing occurred with PU_HV, the self-healing efficiency was 63 and 67%. One specimen performed much worse however (see Fig. 5), a self-healing efficiency of only 18% was found. For this last specimen it was also visually observed at the surface of the specimen that only half of the crack was filled with polyurethane. Crack healing with PU_LV resulted in a sorption coefficient that was lower than the one obtained for uncracked mortar. Consequently, the self-healing efficiency was more than 100%. For the three specimens a self-healing efficiency of 116 to 124% was calculated.

Generally it can be concluded that crack healing with PU_LV resulted in a complete and consistent crack healing for all specimens. PU_HV however was in most cases only able to heal the crack partially, resulting in a wide range of self-healing efficiencies for different specimens.

Conclusions

The presence of cracks has a detrimental influence on the water absorption in mortar. Relatively small cracks (< 100 µm) already caused an increase in the capillary sorption coefficient of 300%. Clearly, repair of cracks will be needed to avoid rapid deterioration of the cementitious material, since water can transport aggressive agents. Therefore, autonomous healing of cracks in cementitious materials by encapsulated healing agents was evaluated in this research for the efficiency of reducing water ingress. As healing agents, two different types of polyurethane were used (one with a relatively low viscosity and one with a relatively high viscosity).

Healing of cracks with the low viscosity polyurethane (PU_LV) resulted in a complete sealing of the crack. The water absorption for these specimens was even lower than for uncracked specimens. A self-healing efficiency of 116 to 124% was found. The main reason for this very high healing efficiency is the fact that quite a lot of healing agent leaked out of the crack and hardened at the mortar surface. One backside of this crack healing is that for an exposure time longer than eight hours, the absorption rate of water started to increase. A possible explanation for this might be the uncomplete healing of the upper part of the crack.

Crack healing efficiency with the high viscosity polyurethane (PU_HV) was much less consistent than healing with PU_LV. Due to the high viscosity of this polyurethane, cracks were not completely healed over the whole crack length. For that reason, a large scatter in the results of the capillary water absorption was found. The maximum self-healing efficiency that was found for crack healing with PU_HV amounted to 67%.

Autonomous self-healing by encapsulated polyurethane was very effective in reducing the ingress of water through cracks in cementitious materials. Especially PU_LV shows very promising results. Consequently, the incorporation of a self-healing mechanism in a cementitious material can be beneficial for the durability of the material and can extend the lifetime of structures.

Acknowledgements

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