Experimental Investigation on the Novel Self-healing Properties of Concrete Mixed with Commercial Bacteria-Based Healing Agent and Crystalline Admixtures

Harry Hermawan[1,2], Virginie Wiktor[3], Pedro Serna[2], and Elke Gruyaert[1](✉)

[1] KU Leuven, Department of Civil Engineering, Materials and Constructions, Ghent Campus, Gebroeders De Smetstraat 1, 9000 Ghent, Belgium
elke.gruyaert@kuleuven.be
[2] Instituto de Ciencia y Tecnología Del Hormigón (ICITECH), Universitat Politècnica de València, Camino de Vera S/n, 46022 Valencia, Spain
[3] Cugla B.V., R&D Center, Rudonk 6b, 4824 AJ Breda, The Netherlands

Abstract. Repairing the cracks in concrete structures is relatively difficult and the manual repair techniques are costly and time-consuming. To overcome this obstacle, stimulated autogenous and autonomous self-healing technologies offer a potential benefit. The healing agents are normally added in the concrete during casting. In fact, traditional concrete also has an autogenous healing ability but the self-healing effect is rather limited. In this study, self-healing concretes were made with two commercial healing agents, namely bacteria-based healing agent (BAC) and crystalline admixtures (CA). The fresh and mechanical properties of concrete were initially evaluated. The addition of healing agents increased the 28 d compressive strength of concrete by 4% for CA and 16% for BAC. The self-healing properties of concrete were evaluated by two methods: (1) crack closure measurements by means of optical microscopy and (2) water flow tests by use of the permeability setup. Results showed that the addition of healing agents showed an advanced progress of the crack closure with increasing healing time, and the permeability rate considerably decreased as a result of the crack clogging by healing products. The self-healing concretes showed better healing and sealing efficiencies than the autogenous self-healing in the traditional concrete, showing a promising result to apply the agents in real applications.

Keywords: self-healing concrete · bacteria · crystalline admixture · self-sealing

1 Introduction

Concrete is relatively prone to cracking due to the high tensile stresses as concrete is considerably weak in tension. The presence of cracks in the concrete is crucial as liquids and gases may enter the cracks slowly jeopardizing the concrete matrix and leading in a lot of cases to corrosion of the steel reinforcement. This phenomenon will shorten the life span of concrete structures and maintenance works will be needed

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to repair the cracks. However, the costs for maintenance are not always low and it requires human intervention. In recent years, thanks to the technological advancements, traditional concrete can be slowly transformed into self-healing concrete. Self-healing concrete involves the inclusion of healing agents in the concrete matrix which help to enhance the healing and sealing performances. This solution is suggested to overcome a limited ability of traditional concrete to autogenously heal the small cracks between 0–100 μm [1]. With the help of specific healing agents, the bigger cracks can be potentially healed. Crystalline admixture (CA) has been used in many studies and the maximum crack width that can be healed with CA varies between 150 and 300 μm [2–5]. Bacterial healing agents show a potential benefit to promote an autonomous healing mechanism. Concrete containing *Sporoscarcina pasteurii* bacteria, cracked to a width of 280 μm, was almost fully healed after being cured for 30 days [6]. The incorporation of *Bacillus subtilis* bacteria was effective to stimulate crack closing up to 520 μm. Other healing agents are also available that may accelerate the healing process such as superabsorbent polymer (SAP) [7, 8], encapsulated healing agents [9, 10], etc. One of the shortcomings in recent development of healing agents is that the healing agents are still often applied in the cementitious paste and mortar, and less effort is done on concrete level. This has been identified by the authors as a research gap. Although it is possible that a better healing or sealing efficiency can be attained by the addition of healing agents into the paste/mortar, it remains rather unclear on how the results observed in the paste/mortar can be transferrable to the concrete. Previous studies [2, 11] by the authors of this paper have identified the effects of the healing agents (crystalline admixture and bacterial healing agent) in the fresh concrete properties. The current paper will continue to understand the effects of two commercial healing agents on the hardened properties of concrete and to assess whether the improvement of healing and sealing properties can be achieved by the introduction of healing agents.

2 Materials and Methods

2.1 Materials

Table 1 shows the mix designs developed in this study. Two commercial healing agents were used, namely Penetron Admix crystalline admixture (CA) and Basilisk bacterial healing agent (BAC). CA is basically composed of Portland cement and proprietary active chemicals, while the detailed composition is kept confidential by the producer. The characteristic of CA is its hydrophilicity, meaning that it easily reacts with water. BAC is mainly composed of dormant bacterial spores from alkaliphilic spore-forming *Bacillus cohnii* which are encapsulated into polylactic acid (PLA) particles and being mixed with other materials such as inorganic salts, calcium lactate (as a nutrient), limestone powder, etc. One of the advantages of this product is that during the healing process, the active microorganism will consume oxygen which will decrease the corrosion process during the process of autonomous crack repair [12]. The recommended dosages of CA and BAC were used in this study at 1% and 2% by mass of cement, respectively. Both healing agents were available in powder form and they were added on top of normal concrete mix. The amount of superplasticizer was adjusted to achieve a similar workability (S4 slump class) for all mixtures. All concrete mixtures were cast with a volume of 70 L and in general, the volume of the mixer was divided into two parts:

- o After mixing the fresh concrete for about 7.5 min, approximately 50 L of fresh concrete was taken out of the mixer. This fresh mixture was partially used to perform the slump and air content tests; and partially cast into nine cubic moulds of 150 × 150 × 150 mm.
- o The remaining mixture of ~20 L in the mixer was continuously mixed and steel fibers (Dramix 35/65) were added in the last stage of mixing at the dosage of 40 kg/m^3 with the purpose of obtaining crack control during splitting of hardened cylindrical specimens. The fresh mix with fibers was mixed for an additional 2–3 min. Finally, the fiber-reinforced concrete (FRC) mixture was poured into six cylindrical moulds of Ø100 × 200 mm.

All specimens were demoulded after 24 h and were cured in the curing chamber with a temperature of 20 °C and RH > 90%.

Table 1. Mix designs

Material	Unit	REF	CA	BAC
CEM III/A 42.5N	kg/m^3	340	340	340
White sand 0/2	kg/m^3	334	333	332
Red sand 0/4	kg/m^3	371	368	366
Limestone 2/6	kg/m^3	239	239	238
Limestone 6/16	kg/m^3	567	567	563
Limestone 16/20	kg/m^3	313	313	311
Effective water	kg/m^3	166	166	166
Effective w/c	–	0.49	0.49	0.49
Superplasticizer	kg/m^3	2.97	2.97	3.86
Penetron Admix CA	kg/m^3	–	3.40	–
Basilisk bacteria	kg/m^3	–	–	6.80
DRAMIX fibres 35/65	kg/m^3	40	40	40
Fresh properties				
Slump	cm	20	21	20
Air content	%	2.5	2.3	2.1
Fresh density	kg/m^3	2393 ± 6	2352 ± 5	2367 ± 4

2.2 Test Setup

The fresh properties of concrete mixtures were evaluated by means of slump, air content and fresh density tests. The mechanical properties were assessed by compressive strength tests at 7, 28 and 91 days. Moreover, the self-healing properties of concrete were assessed by (1) a crack measurement by optical microscopy and (2) a permeability test, both at 0 day (as a reference), 21 days and 121 days after healing. The detailed procedure for the self-healing/self-sealing tests is given below:

- o Each cylindrical specimen (Ø100 × 200 mm) was cut by sawing at 7 days, turning into five discs: one 25-mm top disc, three 50-mm middle discs, and one 25-mm bottom disc. In this study, only the middle discs were used for further tests. These discs were submerged in the water for one day. Next, the discs were cracked by means of a Brazillian splitting test with the target crack width of between 100 and 400 μm. The crack measurement was performed by optical microscope. Per disc, six white marks were indicated along the crack line on two sides (3 marks on the top side, 3 marks on the bottom side). For each white mark, five crack measurements were recorded which later were used to calculate the initial crack width per specimen. In the next stage, after the specimens were healed, the crack measurement was executed again at the same location (see Fig. 1a). The healing efficiency (*HE*) per specimen was calculated as follow:

$$HE = \left(1 - \frac{Final\ crack\ width\ at\ i\ day}{Initial\ crack\ width\ at\ 0\ day}\right) \times 100\% \quad (1)$$

  where *i* is the healing period, either at 21 or 121 days. It should be noted that in a single mixture, there were 6–7 concrete discs. Thus, a global healing efficiency for a single mixture ($\overline{HE}$) was calculated based on the average of *HE*s from all specimens, as follows:

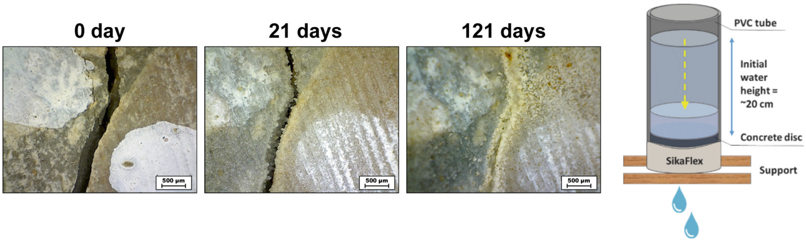
$$\overline{HE} = \frac{\sum_{j=1}^{k} HE_j}{k} \quad (2)$$

  where k is number of concrete discs from a single mixture and j is summation index ($j = 1, 2, 3, \ldots, k$).
- o The permeability test setup is depicted in Fig. 1b. After the crack measurement, the concrete disc was inserted into the PVC tube for approximately half of the height of the disc. Then, to avoid the leaking, the gap between the concrete disc and the tube and the outer surface of the concrete disc were sealed with SikaFlex. After the sealant was dried (for one day), a small amount of water was initally poured in order to check the quality of the sealant and also to make the disc stays in a wet condition. Next, the test was performed by filling the water into the tube and monitoring the reduction of water height from ~20 cm as a function of time, which is later called as a permeability rate. The measurement was done with two repetitions and the average permeability rate was recorded at day 0. After the tests at day 0, the specimens kept in the tubes were fully submerged in the water tanks with a temperature of 20±2 °C in a horizontal position, thus the cracks on the top and bottom surfaces of the specimens were in contact with water. One water tank was used to store all specimens from one mixture, thus in total three tanks were eventually used (to avoid a leaching of healing agent from one mixture to other mixtures). The permeability rate was measured again at 21 days (called here a short healing) and 121 days (called here a long healing).

The sealing efficiency was calculated by Eq. (3) and the global sealing efficiency of a mixture was computed by Eq. (4).

$$SE = \left(1 - \frac{Permeability\ rate\ at\ i\ day}{Permeability\ rate\ at\ 0\ day}\right) \times 100\% \quad (3)$$

$$\overline{SE} = \frac{\sum_{j=1}^{k} SE_j}{k} \quad (4)$$



(a) (b)

Fig. 1. a. Crack measurement by optical microscope, b. Permeability test setup

3 Results and Discussions

3.1 Mechanical Properties

Figure 2 shows the strength development of hardened concretes from 7 to 91 days. At early age of 7 days, no significant difference on compressive strength between the REF and CA or BAC concretes could be detected. At 28 days, the addition of CA increased the strength by 3.6%, but the strength difference between REF and CA concretes is statistically not significant (p-value = 0.62 > 0.05 as a level of significance, tested by t-test). A significant strength difference was found between REF and BAC concretes (p-value = 0.04 < 0.05) with the strength increment by BAC up to 15.8%.

It is also interesting to see that there is a high strength enhancement of BAC concrete from 7 days (48.6 MPa) to 28 days (59.8 MPa). At later age of 91 days, the BAC concrete still maintained the highest compressive strength at 60.9 MPa, followed by CA at 57.0 MPa and REF at 55.0 MPa.

3.2 Self-Healing Properties

Initially, the progress of crack closure was analysed as shown in Fig. 3. After a short healing for 21 days, the REF concrete without healing agents showed three characteristics: (1) most of the cracks were unhealed as shown by the data points close to the bisector line, (2) few cracks were partially healed as shown by the data points below the bisector and (3) the other cracks were fully healed as shown by data points at zero value.

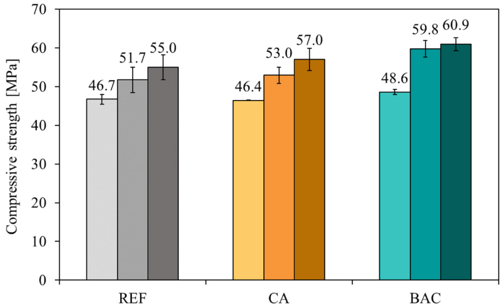


Fig. 2. Compressive strengths of all concrete mixtures (note: for each mix, the left, middle and right bars represent strength values at 7, 28 and 91 days, respectively)

As compared with REF, it is clear that the addition of healing agents showed advanced progress on the crack closure. This notion is shown by the fact that the data points from CA and BAC mixtures are scattered below the bisector, meaning that the initial crack width becomes narrower as a result of the formation of healing products on the crack tip. On the other hand, some cracks were also observed to be already fully healed. After a long healing for 121 days, most of cracks were completely healed as the points mostly lie at the x-axis. Only a few data points were still seen below the bisector and none on the bisector line.

In order to quantify the healing efficiency of concrete mixtures, data obtained from Fig. 3 were analysed by categorizing the crack sizes into four ranges including 0–100, 100–200, 200–300 and 300–400 μm. Per crack width range, all data points were taken to calculate the healing efficiency by Eq. (1) and the results were shown in Fig. 4. Small cracks with the size below 100 μm, regardless of the addition of CA or BAC, were always fully healed with 100% *HE* after 21 and 121 days of healing. This finding is supported by the fact that the autogenous healing mechanism normally works best with the small crack width as reported by [1]. A large variability of *HE* results was seen for the cracks larger than 100 μm. This may occur due to the different crack patterns on all specimens. In fact, this is unavoidable considering difficulties in controlling the crack width during the splitting test and the random selection of measuring points along a crack. To better analyze the results, a series of statistical analyses was conducted. Shapiro-Wilk test and Levene's test were initially conducted and the results confirmed that the data set did not follow a normal distribution and the variances were not equal. Thus, ANOVA test by Kruskal-Wallis test was selected to analyze this type of data. Nevertheless, the groups of data among 100–200 and 200–300 μm, regardless of the healing time, did not show any significant differences (p-values > 0.05). In fact, a significant difference was found on the healing efficiencies between REF–CA (p-value $= 0.015 < 0.05$) and also REF–BAC (p-value $= 0.004 < 0.05$) for large cracks in the range of 300–400 μm. The global healing efficiency was further calculated by Eq. (2) based on all obtained data from Fig. 4, and the results were summarized in Fig. 5a. The mean $\overline{HE}$ results at 21 days were recorded at 61, 69 and 73% for REF, CA and BAC, respectively. The trend of healing efficiency at 121 days also follows the result from 21 days and both healing agents slightly showed a beneficial effect. Although a high variability and no significant

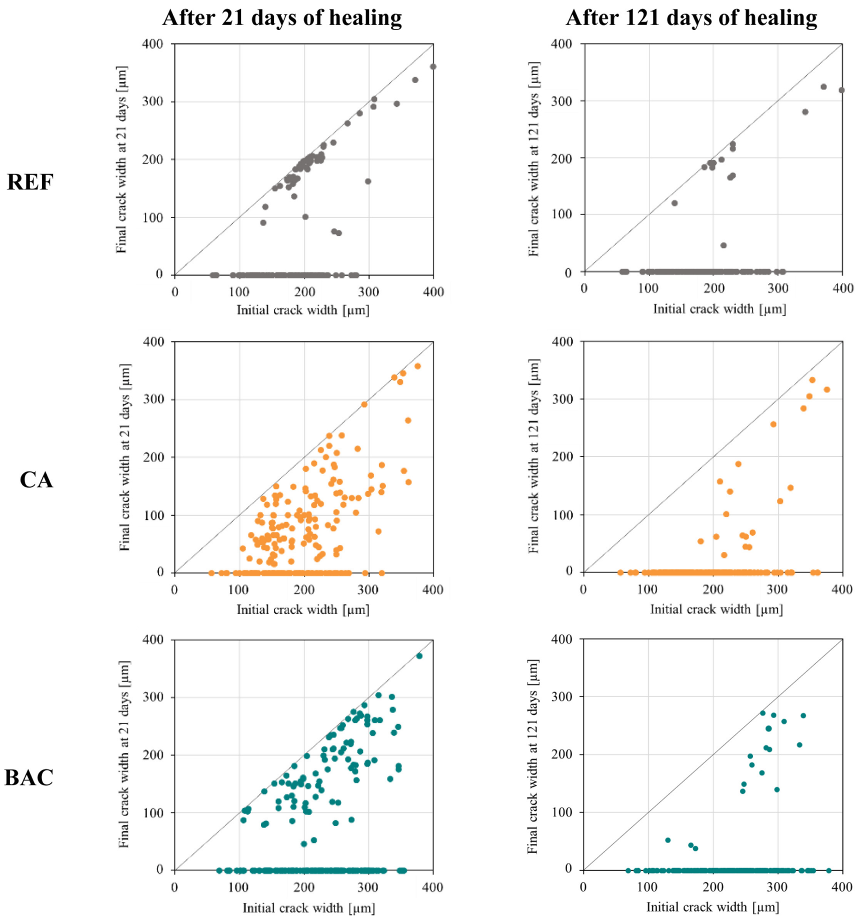


Fig. 3. Progress of crack closure on one measurement after being subjected to 21 and 121 days of healing

difference as previously seen, the current trend seems to indicate that the addition of healing agents can help to slightly enhance the healing efficiency. This effect is more clear for wider cracks. In addition, the results seem more stable with lower variation with the introduction of healing agents as compared with REF. This result gives an important clue that an optimization of mix designs for self-healing concrete is necessary to further improve the self-healing effects.

3.3 Self-Sealing Properties

To confirm the sealing ability of the concrete mixed with different healing agents, permeability tests were conducted. For each mixture, there were 6–7 concrete discs and each disc was tested by the permeability test. In this regard, the permeability rate was derived from the water height reduction over time, as shown in Fig. 6 with BAC mixture as an

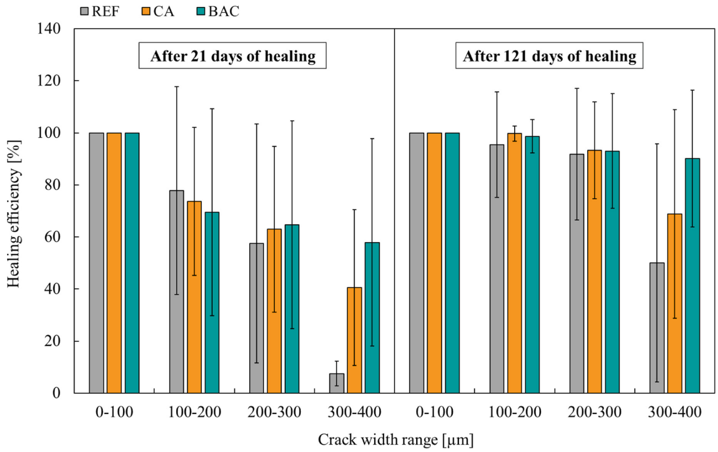


Fig. 4. Healing efficiency of concrete mixtures depending on the crack sizes (note: the numbers of measures for each crack size group of [0–100, 100–200, 200–300, 300–400 μm] are [8, 104, 55, 5] for REF, [6, 108, 76, 14] for CA, and [5, 66, 113, 23] for BAC)

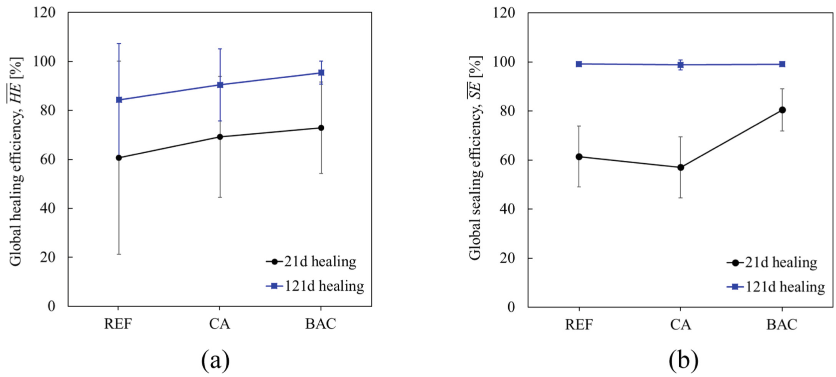


Fig. 5. a. Global healing efficiency and b. Global sealing efficiency of different concrete mixtures

example. It is clear that after the specimens were cracked and subsequently tested by the permeability test, the water level dropped rapidly through the crack as the slopes had steep inclinations. It is noteworthy to mention that there is a high variability in permeability results which is attributed due to the variety of crack width. The cracked specimens were not differentiated based on the crack width range and a mix of cracks between 100 and 400 μm was available in all tested specimens, which eventually may affect the permeability results. However, after healing for 21 days, there were good improvements as the reductions of water were relatively slow, as compared with the initial values. This means that the cracks became narrower after healing due to the formation of healing products inside the crack. However, the cracks were not completely closed in this scenario as in some specimens, the water reductions were still apparent. With a longer healing time up to 121 days, the water level remained constant, meaning no further reduction of water height that indicates a 100% sealing efficiency. The global sealing efficiency ($\overline{SE}$) was calculated by Eq. (4) and the results were summarized in Fig. 5b. The addition of CA apparently did not show a considerable improvement of sealing capability as compared with REF. However, the BAC showed a significant improvement with approximately 80% $\overline{SE}$ by average after a short healing or around 30% sealing improvement as compared with REF. This result may be associated with a great formation of healing products inside the cracks. It should be noted that the crack measurement by optical microscope was done on the crack tip and the crack closure inside the crack could not be measured by this instrument. Thus, this permeability test can indirectly "sense" the presence of healing products inside the crack which is presented by a reduction of the rate of water permeability. At a relatively short healing, the addition of BAC is more effective than CA. Moreover, after a long healing, the $\overline{SE}$ results were identical for all mixtures regardless of the addition of healing agents. This might be explained by the fact that a 121 healing days is considered a very long period to seal the cracks, thus the effect of healing agent is not obvious. In contrast, when the specimens were subjected to a short healing, there was an improvement of sealing by healing agents. It may suggest that a further study is necessary to regularly monitor the progress of sealing effect as increasing healing time before 4 months (~121 days) (Fig. 6).

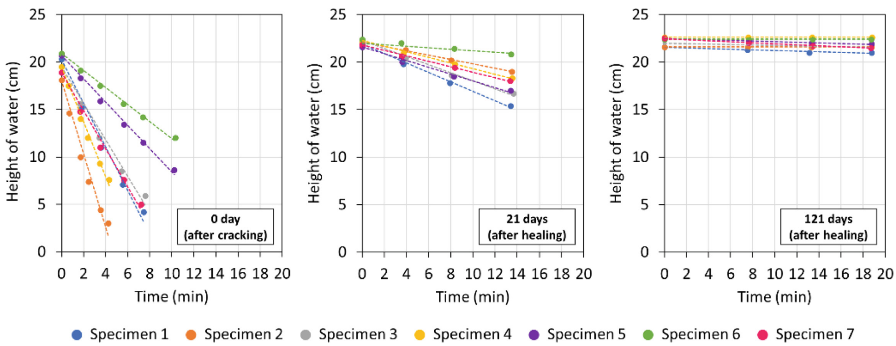


Fig. 6. Permeability rate (BAC mixture taken as an example)

3.4 Analysis of Healing Products

An additional study was conducted to investigate the polymorph of healing products and whether the addition of healing agents will alter this formation. The healing products from each mixture were collected by carefully scraping the precipitates near the crack tip, so not at the inside of the crack (see Fig. 7). In this case, the analyzed products were always superficial. Next, a Fourier transform infrared (FTIR) spectroscopy was employed to analyse the composition of the self-healing products. The results were generated in the spectra bands as shown in Fig. 8. Apparently, there is a high degree of similarities in FTIR spectra of all samples taken from different mixtures. The major peaks of calcium carbonate ($CaCO_3$) were characterized at the bands around 1449–1467 and 854 cm^{-1} [13, 14]. According to literature, $CaCO_3$ in the form of aragonite displays a characteristic symmetric carbonate stretching vibration at 1081–1087 cm^{-1}, a carbonate out-of-plane bending vibration at 844–858 cm^{-1}, and a double degenerate planar bending at 712–713 cm^{-1} in its FTIR spectrum [15–17], while $CaCO_3$ in the form of calcite is mainly characterized in the out of bending plane at 873–878 cm^{-1} and doubly degenerate planar bending around 712–713 cm^{-1} [15–18]. Eventually, the mineral coming outside from the crack are confirmed to be calcium carbonates in the polymorph of aragonite.

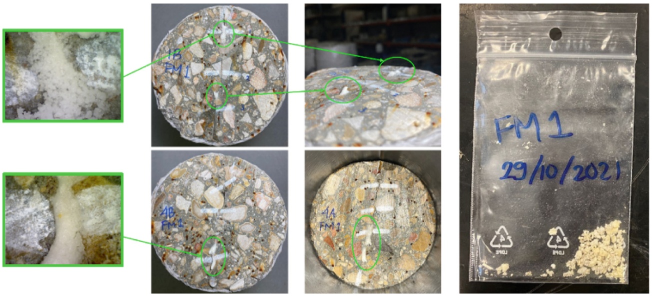


Fig. 7. Collecting the healing products from the crack tip

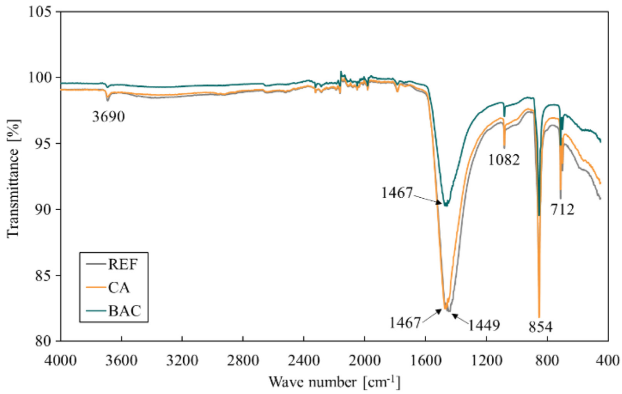


Fig. 8. FTIR analysis of healing products

4 Conclusions

This paper aims to evaluate the effect of incorporating healing agents into the concrete with regard to the mechanical, self-healing and self-sealing properties. Penetron Admix crystalline admixture (CA) and Basilisk healing agent (BAC) were used as healing agents with the dosages of 1% and 2% by cement mass, respectively. For self-healing purpose, the 7-days old concretes were cracked and subsequently healed for 0, 21 and 121 days under full water immersion. Based on the obtained results, the following conclusions can be drawn below:

1. The highest increment of 28 d compressive strength was achieved by BAC concrete with 15.8% improvement, while the addition of CA slightly increased the strength by 3.6%.
2. The addition of healing agents accelerated the healing progress at a relatively short healing time. Most of the cracks were completely healed with a longer healing time regardless of the inclusion of any healing agents.
3. Based on the 21 and 121 days of healing, both CA and BAC showcased a small improvement on the healing efficiency and a lower variability of the results can be achieved as compared with REF. In terms of sealing efficiency after 21 days of healing, the introduction of BAC had a better improvement than REF and CA. The influence of BAC is more important for the wider cracks (300–400 µm). Furthermore, when the healing time was prolonged to 121 days, all concretes were almost fully sealed (~100% sealing efficiency) regardless the inclusion of any healing agents.
4. Calcium carbonates in the polymorph of aragonite were confirmed to be the mineral coming outside from the crack.

Based on these findings, it is suggested for future studies to look into the modification of self-healing concrete mix designs with the aim to further enhance the healing and sealing abilities.

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