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Bio-Based Self-Healing Mortar

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Abstract: This paper investigates the effects of alkaliphilic spore-forming bacteria of the genus Bacillus on the compressive strength of the mortar cube and the healing capacity of the bacteria as healing agent on mortar containing crack. The experiments were carried out using cube test, stereomicroscopy and environmental scanning electron microscopy (ESEM). Cracked mortar specimen with and without the presence of the bacteria-healing agent were prepared. Results showed that the inclusion of these bio-healing agent in mortar mixtures cause more pore volume, which has no significant effect on compressive strength development at 28 days. In the series of healing capacity test, the cracks were significantly healed in bacteria-based than in control specimens after 28 days incubated in water bath and thus, increase permeability resistance of bacteria-based mortar specimens.

Keywords: Self-healing, crack, mortar, bio-healing agent

1. Introduction

Deteriorating agents such as chloride, carbon dioxide, oxygen, sulphate and other potential materials can be linked to the micro-cracks on cementitious materials and also to specific materials properties such as porosity. In consequence, the degradation processes may accelerate and affect the durability in further. With respect to durability improvement, self-healing materials target sealing of occurring cracks via deposition of crack-filling material which promotes a sustainable repair methodology resulting in an increase in lifespan of concrete structures [1].

Bacteria-based self-healing agent has a great potential to provide self-healing capabilities to cementitious materials [2]. This technique has been studied by Ramachandran et al., 2001 [3] with calcite deposition induced by Bacillus pasteurii using various concentrations added into cement mortar cubes containing simulated cracks of different depths. The sealing in shallower cracks was observed to be more efficient in comparison to deeper cracks due to active growth of bacteria by the presence of oxygen. Bang et al., 2001 [4] had added the use of polyurethane (PU) to immobilize the whole cell of Bacillus pasteurii and found PU as an effective improvement method in microbiologically-induced calcite precipitation (MICP) in concrete cracks. Both authors concluded that the MICP technique was effective to remediate cracks in building materials. The conclusion was in good accordance with studies done by Achal et al.,2010, 2013 [5, 6], whose observed positive effect of Bacillus sp. CT-5 on the mortar compressive strength and permeation properties. The authors had also examined the calcite production ability of Sporosarcina pasteurii that can produce high urease activity. As a result, a significant improvement in surface permeability resistance was observed [7].

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In Belgium, bacteria or derived ureolytic were externally applied on degraded limestone and cement-based materials as surface protection for reducing the water absorption [8-10]. The bacteria of the *Bacillus sphaericus* and *Bacillus lentus* were examined for calcite deposition on limestone through the process of ureolytic calcium precipitation. The hydrolysis of urea into carbonate and ammonium is catalysed using urease. By the deposition of carbonate layer on the surface, the establishment of a biofilm was initially proposed in the study by Dick et al. [8]. Further investigation of biodeposition treatment on durability aspects was studied by De Muynck et al. [10]. With a different treatment procedure applied, the biodeposition treatment resulted in an increased resistance of mortar specimens towards degradation processes.

The newly developed bacteria-based self-healing agent [11] using alkali-resistant spore-forming species of the genus Bacillus and more specifically related to the species Bacillus pseudofirmus and Bacillus cohnii was proven to have the same advantages of MICP technique. These types of bacteria which can also survive in high temperature conditions could act as an autonomous repair system in which, an active crack remediation is done by the hardened mortar or concrete with the aid of bacteria. The bacteria work optimally in a pH range of 8 to 11.5 and in a temperature range of 15 – 40 °C. If untouched, the bacterial spores in the healing agent can potentially stay inactive in the concrete for centuries [12].

On the other hand, under suitable environmental conditions with appropriate food source available and sufficient water, the bacteria can grow exponentially and heal the cracks [22]. Therefore, when water begins to seep into the cracks it activates the spores of the bacteria which subsequently germinate into active (vegetative) cells which metabolically convert the calcium lactate. This process consumes oxygen and results in the formation of limestone that solidifies and seals pores and cracks. In general, the bio-conversion of calcium-lactate results in direct calcium carbonate ($CaCO_3$) formation according to the following reaction:

$$CaC_6H_{10}O_6 + 6O_2 \rightarrow CaCo_3 + 5CO_2 + 5H_2O \quad (1)$$

The metabolically produced CO_2 can subsequently chemically react with calcium hydroxide present in the paste matrix leading to additional or indirect $CaCO_3$ precipitation, according to:

$$5CO_2 + 5Ca(OH)_2 \rightarrow 5CaCO_3 + 5H_2O \quad (2)$$

These bacteria spores are quite similar with plant seeds. They are separated by a very thick cell wall which makes them well-resisted against deficiency, aggressive chemicals or extreme mechanical loads. Like a seed, the spore will patiently wait for the suitable environment to grow. Under favourable conditions, the bacteria will be activated from its dormant state to germinate and will be distributed again. In this way, the drawback of urea hydrolysis by formation of ammonia which results in excessive environmental nitrogen loading can be avoided [1, 13].

2. Materials and Methods

2.1 Selection of Bio-Healing Agent

The alkaliphilic spore-forming bacteria of the genus *Bacillus* used in this study were obtained from the German Collection of Microorganisms and Cell Cultures (DSMZ), Braunschweig, Germany and cultivated spores were mixed with calcium lactate, acting as mineral precursor compound, and other growth requiring nutrients before incorporated into the concrete mixture [1, 14]. The bacterial spores and nutrients were protected by encapsulation using a binder (stearic acid) to increase the potential for long-term viability of bacterial spores.

The bio-healing particle was prepared using powder compression technique. The dried bacterial spores and nutrients (both in freely flowing powder form) were mixed with 1% (weight) binder. This form of compressed powder (cylindrical-shaped) healing agent with uniform size is hard and consists of almost fully (95 – 99%) functional healing agent components (Fig. 1). Therefore, the applied volume of healing agent can be limited to only 1% of concrete volume and 4% of cement weight [15]. This two-compound healing agent, acting as a healing particle, was embedded in the concrete or mortar and thus became an integral part of the material.

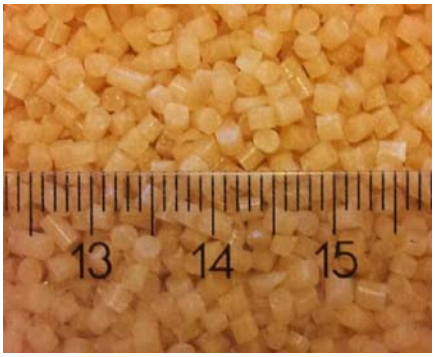


Fig. 1 - Cylinder-shaped healing agent particles with uniform size added to the mortar mix

2.2 Preparation of Samples

Standard mortar prisms of dimensions 40 mm x 40 mm x 160 mm made from ordinary Portland cement (CEM I 42.5), CEN Standard Sand and 0.5 water to cement ratio in accordance with EN196-1:2005 without any further additions (control specimens, SM) and with bacteria-based healing particles (BM) were prepared. Table 1 summarizes the composition of each batch required for production of three replicate test specimens with a quantity of 15 kg healing agent per cubic meter of mortar was applied. Six mortar cubes were tested for compressive strength after 1, 3, 7, 14 and 28 days curing and six mortar prisms in each group of SM and BM were prepared for healing capacity test after 18 weeks. The mortar prisms were left for 72 hours curing at room temperature before being unmoulded. Afterward, all samples were further cured in a fog room at 99% ± 1% relative humidity until 28 days.

Table 1 - Composition of mortar mix required for three test specimens.

Compound	Weight (g)
Cement (CEM I, 42.5)	450
CEN Standard Sand	1350
Water	225
Healing agent	13

2.3 Pre-Conditioned of Samples

three-point bending using an Instron machine of 10 kN maximum capacity (as shown in Fig. 2). Prior to the three-point bending test, five sides including the as-cast surface of the mortar sample were sealed with the aluminum sticker. The crack width was measured using a linear variable differential transformer (LVDT). Under crack displacement control at a speed rate of 0.5 µm/s, a visible crack with unloaded crack openings of approximately 350 µm were produced to investigate self-healing potential bacteria-based specimens.

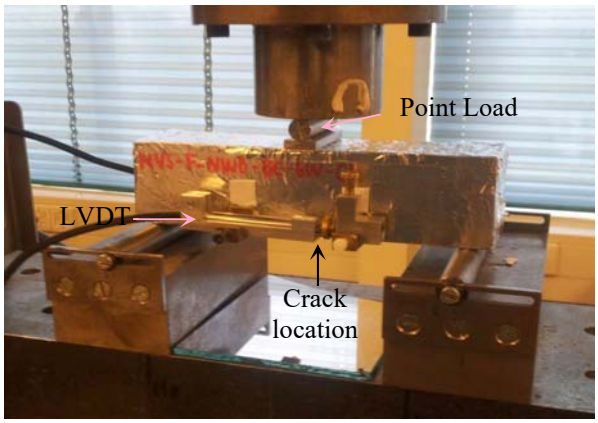


Fig. 2 - Three-point bending test

The specimens were reinforced with two steel wires at the tension side in order to keep the crack surfaces at a distance after unloading and to avoid full separation of prism half during the bending test. The support span of the three-point bending test was 142 mm and the loading span was 40 mm. The load and crack displacement were recorded during the tests. After the maximum crack open width was reached, the load was released.

The crack line images were taken by using a stereomicroscope. Immediately after the surface cracks were recorded, the specimens were fitted into tray and partially submerged with crack opening side facing down into tap water. Specimens with and without bacteria-based healing agent were kept in separate containers to avoid cross-infection [17].

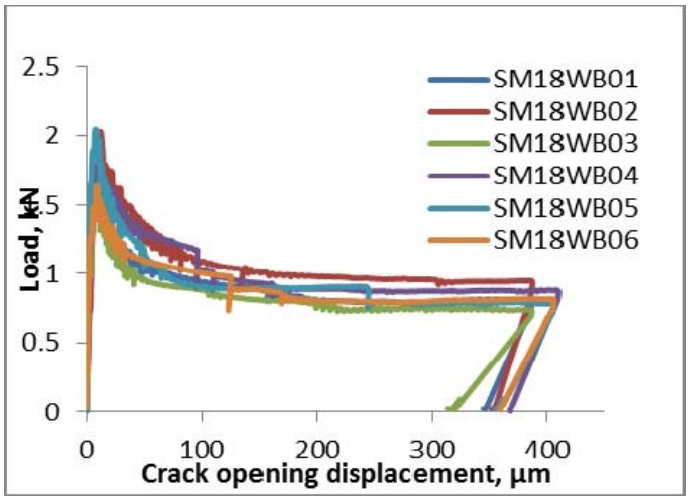
2.4 Optical Observation by ESEM and Stereomicroscopy

Measurements were carried out to conclude that the enhanced healing performance in the bacterial specimens can be attributed to the bacteria-based healing system. The surface crack was evaluated with a stereomicroscope to investigate the surface crack closure before and after the healing treatment and also after the exposure to the test environments. More detailed information on the crack healing was obtained by observing the mineral precipitates along the crack surface by ESEM. The specimen was examined under ESEM using magnification between 20 to 40 times to observe for calcium carbonate precipitation. The observed mineral formation was mapped on the enlarged image.

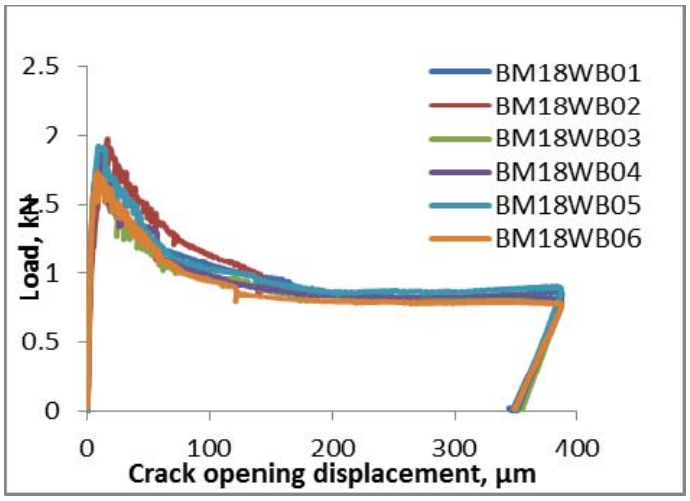
3. Results and Discussion

3.1 Crack Opening Displacement

The crack width was kept in constant range to limit effect from crack width on the ingress (Achal et al., 2013). A single crack appeared approximately in the middle of the prism with a span of 142 mm. The load displacement graph is given in Fig. 3(a) and Fig. 3(b) for SM and BM specimens respectively. The crack produced from 3-point bending test was not straight but appeared more tortuous. The visible crack length from the side of the specimens was roughly 60 mm to 90 mm. From the graph, it can be seen that most SM specimen were cracked between 320 µm to 350 µm displacement after unloading. However, more consistent crack opening displacement of 350 µm was recorded for BM specimens.



(a)



(b)

Fig. 3 - Load-displacement curves of (a) standard mortar (SM) and (b) bacteria mortar (BM) specimens.

3.2 Compressive Strength Development

Fig. 4 compares the effect of healing agent addition on strength development between the mortar cubes with and without healing agent. The incorporation of healing agent in mortar specimens resulted in a positive development of compressive strength. Mortar strength of healing agent amended specimens was lower in 1, 3, 7 and 14 days cured specimens, it was respectively equal and 7% higher in 21 and 28 days cured specimens. Apparently compressive strength of healing agent amended specimens overtakes that of reference specimens at later age.

The development in strength of mortar samples amended with healing agent can possibly be explained by the lower healing agent to cement ratio (2.9% healing agent concentration) and additionally by the lower surface to volume ratio of the cylindrical healing agent applied to the mortar mix. Both effects apparently result in less amount of organic compound (calcium lactate) release from the healing agent particles during the mixing process, and therefore

reduced negative effect on compressive strength development. Also, in another study an actual increase in compressive strength of older (28 days cured) specimens was found for healing agent to cement weight ratio of around 3% (Renee Mors, personal communication). In addition to that, a smaller size of healing particles may contribute to a more even dispersion of particles in the cement matrix what can possibly improve crack healing efficiency of specimens. In some cases it was observed that the added calcium lactate not only acts as precursor compound for crack sealing minerals but also enhances concrete compressive strength [14].

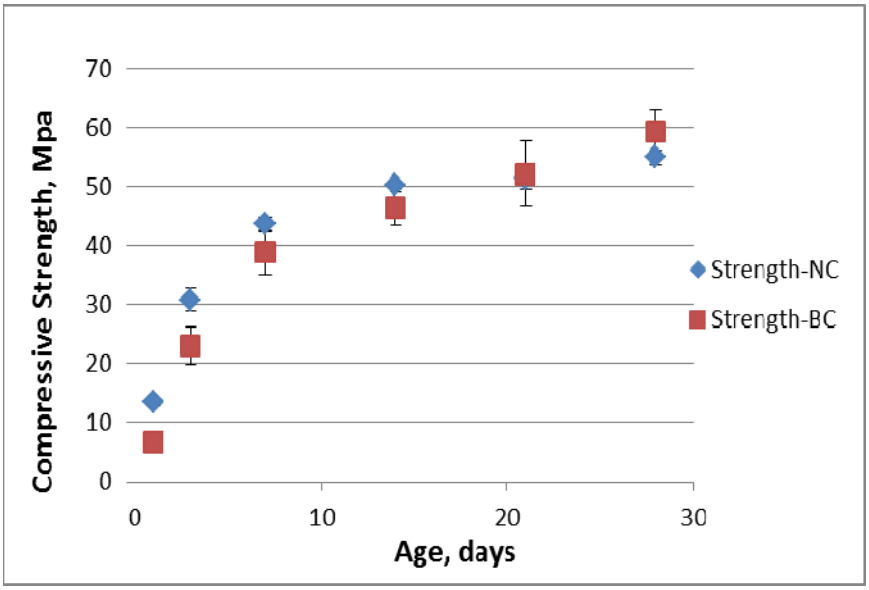


Fig. 4 - Compressive strength development of mortar without (NC) and with bacterial healing agent addition (BC)

3.3 Crack Opening Displacement

Cracks closure at the crack mouth from both control and healing agent-specimen were visualized by stereomicroscopy. Fig. 5 shows images taken immediately after three-point bending test and after 28 days of healing treatment of healing-agent specimen. It was observed that the autonomic-healing already took place as mineral formation was clearly visible at the crack mouth in bacteria-based specimen. In a study by Wiktor et al. [4], it was observed that complete crack-sealing in bio-based concrete took 100 days of immersion in tap water for 460 µm wide cracks. While in this case at least 50 % of 350 µm (mean value) wide cracks were physically blocked in bio-healing agent containing samples due to difference in period of healing treatment. The 50 % was qualitatively measured by considering the length of crack filled (sealed) with mineral to the unfilled (unsealed) crack length.

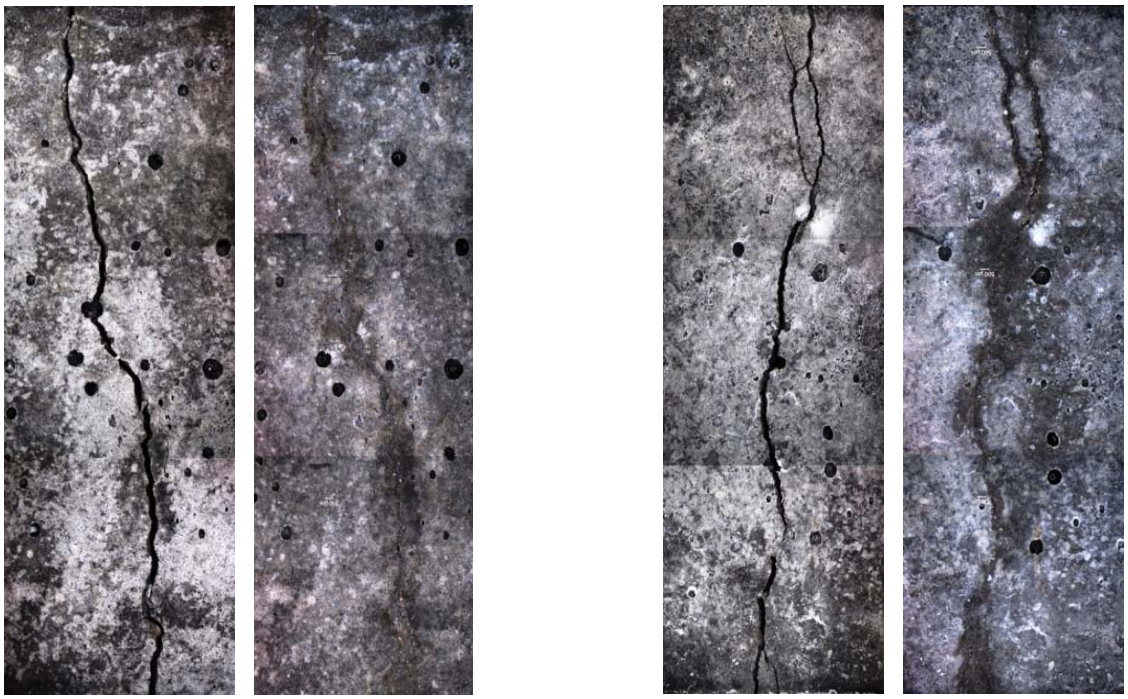
In the case of healing by bacteria, calcium carbonate precipitation could be attributed to physicochemical environmental conditions for metabolic activity. For instance, limitations of minerals availability or too high or too low pH are factors which influence the bacterial calcium carbonate precipitation rate. In addition to that, using tap water was proven to increase healing efficiency in comparison with using demineralized water in healing treatment. Therefore, repetitive replenishment of the specimen incubation water by fresh oxygenated water could increase the rate of bacterially controlled crack sealing.

In contrast, no obvious mineral formation, i.e. autogenous healing, was detected on the crack surface of control specimen as shown in Fig. 6. Moreover, less than 50 % of the similarly sized cracks in control samples were visually sealed after 28 days immersed in demineralized water. As a matter of fact, an autogenous healing was only effective for crack widths of less than 200 µm [18, 19].

From these it could be concluded that at least 50 % of healing occurred in bacteria-based specimens at the end of four weeks treatment in wet conditions. From these observations it can be concluded that the transportation of an aggressive agent will be delayed due to blocking of the cracks, and that this effect is significantly more pronounced in specimen amended with bacteria-based healing agent.

The healing occurrence was further observed by the presence of whitish substance deposits along the internal crack line of a few impregnated samples with the healing treatment process as shown in Fig. 7(a) and Fig. 7(b). The area with the whitish substance was scratched to make it exposed to the surface of the thin epoxy layer for element quantification by EDS point analysis. The analysis was carried out in three distinctive areas to clearly distinguish the peak of the mineral formation. From the results given in Fig. 8(a) and Fig. 8(b), point one (1) and three (3) exhibit higher peak counts for calcium and carbonates. While point two (2) indicates that the area was still covered by a thin layer of epoxy.

Qualitatively, it indicates a weak appearance of C-S-H products at the spotted area. Moreover, it was also reported the weight % of oxygen was three times of calcium which signify the minerals formed was calcium carbonates based.

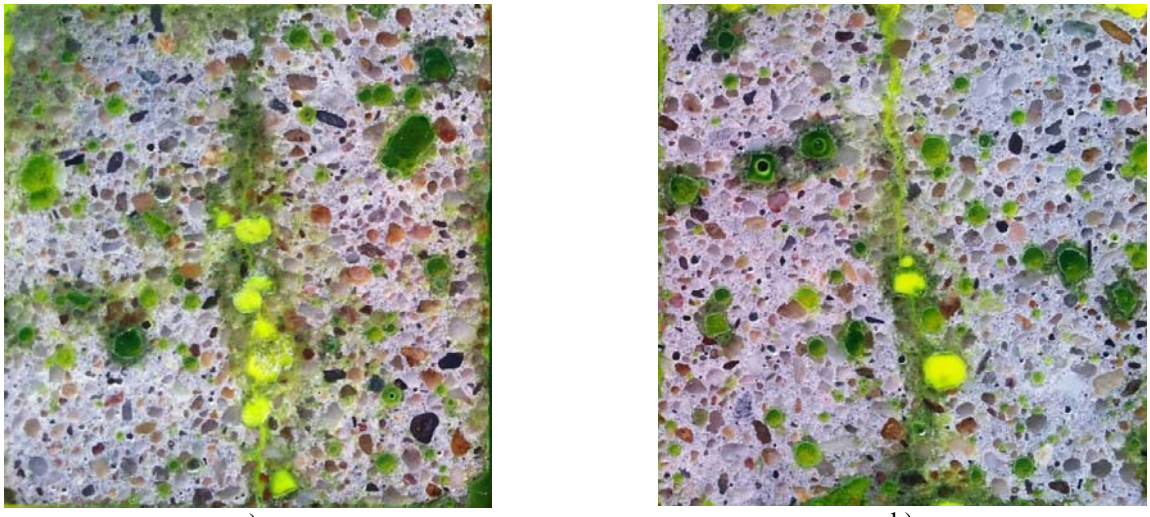


a) After bending test b) 28 days healing

Fig. 5 - Visualization of crack-healing process by stereomicroscopy before and after healing treatment for bacteria-based specimens.

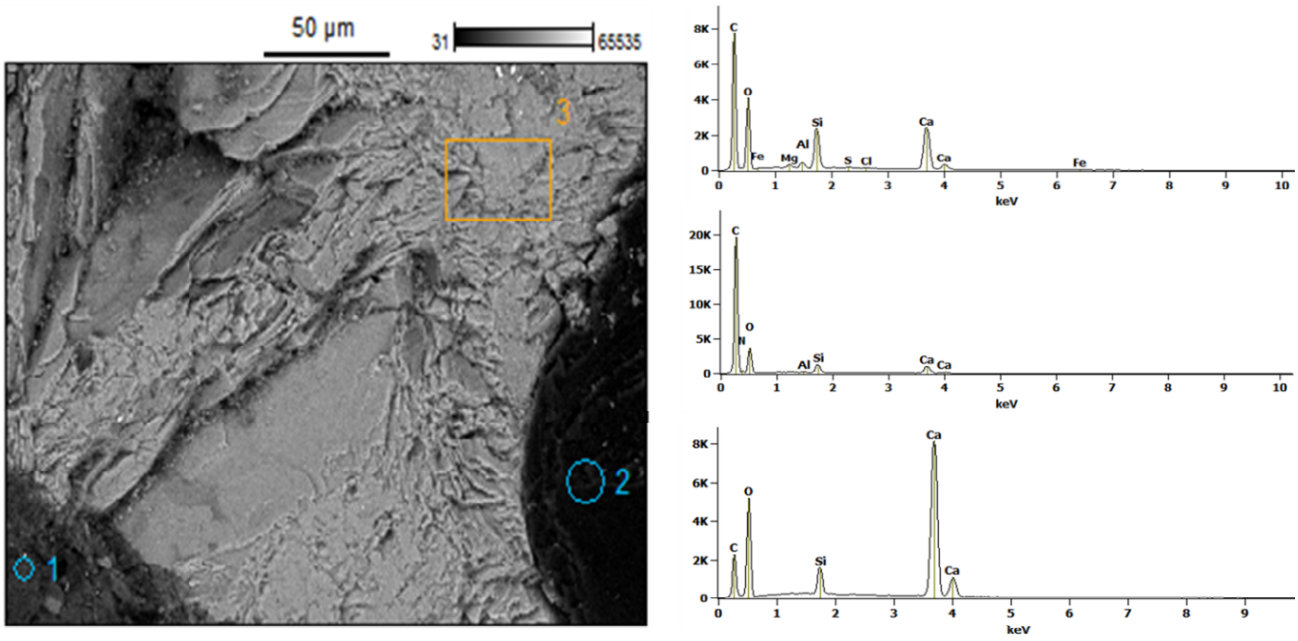
a) After bending test b) 28 days healing

Fig. 6 - Observed of crack-healing process before and after healing treatment in control specimens by stereomicroscopy.



a) b)

Fig. 7 - Evidence of healing spots (a) by the presence of whitish deposits along the internal crack line with (b) indication of scratched area for EDS point analysis of epoxy impregnated samples.



a) b)

Fig. 8 - EDS point analysis (a) in three different regions with (b) element counts.

4. Conclusion

The healing capacity of bacteria-based healing agent on mortar containing crack was investigated using techniques of stereomicroscopy and scanning electron microscopy. The compressive strength development of concrete and mortar specimens with incorporated bacteria-based healing agent was also characterized in this study. Based on the results presented, it was concluded that the inclusion of these bio-healing agent in cementitious mixtures cause more pore volume, which has no significant effect on compressive strength development at 28 days. In addition to that, 4-week healing treatment showed the crack was almost filled with calcium carbonate in specimen of demineralized water solution-incubated bacteria-based mortar specimens. In contrast, no obvious of calcium carbonate precipitation was detected on the crack surface of control specimen. Based on the present study it can be concluded that crack sealing was more substantial in bacteria-based longer incubated specimen in comparison to equally long incubated control specimen.

The results have also shown that cracks in bacteria-based specimens containing system were sealed through the process metabolically mediated calcium carbonate precipitation with proof-of-principle observed in the enhancement

sealing deposit. Crack filling with mineral precipitates was also observed by the presence of the whitish substance deposits along the internal crack line which was not observed in control specimen.

From the results, it is strongly suggested that healing particles are existence in hardened matrices with metabolically active bacteria and nutrient available in time. It was concluded that, although in a cracking event with the presence of water, oxygen, and nutrients, the bacteria can remain dormant if the values of the temperature and pH are not compatible with their metabolic capacity. Therefore, a specific combination of a metabolic pathway, activity, and physicochemical environmental conditions are essential for bacterial metabolically driven calcium carbonate precipitation.

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