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BOND ENHANCEMENT OF REPAIR MORTAR VIA BIODEPOSITION

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Abstract

The bond between repair mortars and existing concrete substrates is critical for the long-term performance and durability of the repaired structure. The carbonation state of the substrate is one of the parameters that may affect this bond strength. The type of calcium carbonate polymorph (calcite, aragonite or vaterite) affects the nucleation and growth of cement hydration products. In this study, carbonation of the mortar substrate is promoted via the biodeposition of calcium carbonate by a ureolytic bacteria strain previously employed in bioconsolidation. X-ray diffraction and scanning electron microscopy were used to examine the interfaces between the repair material and the substrate, as well as the polymorph of the deposited calcium carbonate. It appeared that the approximately 50 µm thick biodeposition film on the mortar surface mostly consisted of calcite and vaterite. The deposited crystals were full of bacterial imprints. Both the repair material and the substrate tended to show a good adherence to that layer. The bond, as assessed in this study by slant shear specimen testing, was improved by the presence of the biodeposition layer.

1. INTRODUCTION

Concrete is one of the most widely used construction materials on Earth. It is an ideal material to resist compressive forces, but when sufficient tensile forces are present, the restrained concrete may crack. And, without repair of the cracks, the durability can be critically compromised. One can decide to use a self-healing concrete during the design phase of construction [1-3], but repair of existing concrete structures will still often be needed. This manual repair should be made with care and precautions should be taken to assure that the repair is long-lasting, durable and efficient. If the bond between repair product and concrete substrate is not sufficient, delamination or spalling may occur. Therefore, one needs to make sure that the surface treatment of the substrate is properly executed.

One way of increasing and controlling the bond between the repair material and the concrete substrate could be the use of a biodeposition layer. One of the first patented applications on biodeposition was the protection of ornamental stone by a microbially deposited carbonate layer [4, 5]. This calcium carbonate precipitation can be biologically induced by bacteria. These bacteria synthesize the minerals in a unique form, characteristic of the specific species of bacteria. This biodeposition technique has also been applied on cementitious materials resulting in an increased resistance of mortar specimens towards chloride penetration, freeze/thawing and carbonation [4, 6-8]. It should merely be considered as a coating system as carbonate precipitation is mainly a surface-controlled phenomenon due

to the limited penetration of bacteria into the porous cementitious matrix. Thin-section analysis revealed that the thickness of the bacterial layer was typically within the range of 10 µm to 40 µm; in which larger crystals up to 110 µm could be found [4]. This layer can be a promising route to engineer the substrate surface for optimal bond strength characteristics.

In this paper, the bond strength was assessed by slant shear testing. Specimens with and without biodeposition layer were studied and the crystal formation composition and morphology were examined.

2. MATERIALS AND METHODS

2.1 Bacterial strain and cultivation condition

Bacillus sphaericus LMG 22257 (Belgian coordinated collection of microorganisms, Ghent) was used in this study. The bacteria were grown aseptically in the growth medium (400 mL per batch) which consisted of a blend of yeast extract (20 g/L) and urea (20 g/L). The culture was incubated at 28 °C on a shaker at 100 rpm [10.5 rad/s] for 24 h. Subsequently, the bacterial cells were harvested by centrifugation (7000 rpm [733.0 rad/s], 7 min) of the 24 h old grown culture and were resuspended in sterile saline solution (NaCl, 8.5 g/L). The concentration of the bacteria in the suspension was $1.5 \cdot 10^9$ cells/mL to $2 \cdot 10^9$ cells/mL. The obtained bacterial suspension was stored in a 4 °C refrigerator for further experimental use.

2.2 Mortar specimens

The standard used to make the mortar substrates was ASTM C882/C882M-13 on 'Bond strength of epoxy-resin systems used with concrete by slant shear'. Six Portland-cement mortar cylinders with a standard mixture composition according to EN 196-1 were cast (510 kg/m³ CEM I 52.5 N, 1530 kg/m³ silica sand 0/2, and 255 kg/m³ water). The specimens' diameter and height were 75 mm and 150 mm, respectively, and each had a diagonally cast bonding area at a 30° angle from the vertical, as per the ASTM standard.

The specimens were manually ground (bonded surface) by means of a sand paper until the desired roughness was reached. The International Concrete Repair Institute (ICRI) has a set of "roughness" surface profile chips (http://www.jdtechnical.com/Surface_Prep-JD.htm). An intermediate profile, similar to the CSP-5 chip, was targeted at an age of 28 d. All prepared surfaces were visually similar.

2.3 Biodeposition treatment

Mortar specimens (BAC) were partially immersed in a precipitation medium which consisted of urea (30 g/L), calcium nitrate (118 g/L) and yeast extract (5 g/L) for 24 h. The medium level was approximately 10 mm above the immersed surface (elliptical surface for applying repair material) of the mortar specimens. After that, the specimens were taken out from the precipitation medium and put upside down until surface dry. Subsequently, bacterial suspension was sprayed (approximately 0.5 mL/cm²) all over each elliptical surface every 6 h for 4 times.

2.4 Repair material application and slant shear testing at 28 d

All mortar substrate specimens were soaked in tap water for 24 h. The face of the mortar section was put on an absorbent material for 10 min and was subsequently dried in air for 15 min. The repair material (Sika MonoTop-124 N[1]) was mixed for 3 min. The prepared bonding surface was put horizontal and 2 mm of repair material was applied, followed by the application of a second mortar specimen. The entire cylinder (especially the sides) was(/were) covered with plastic foil and tape to exclude movement during hardening of the repair material. The specimens were kept horizontal for 48 h. Afterwards they were put in a moist room at 95 ± 5% RH and 20 ± 2°C until the repair product achieved an age of 28 d. Prior to testing, the loading surfaces of each cylinder were ground to produce a smooth testing surface. The composite specimen was loaded in compression and its strength was recorded. The bond strength was determined by dividing the load carried by the specimen at failure by the area of the bonded surface. The area of the bonded surface was reduced by that of any voids found in the bond layer on inspection after the test. Only voids larger than 3 mm in diameter were taken into account.

2.5 Characterization of the interface between mortar substrate and repair material

The mineral phases in the mortar-biodeposition layer-repair material interface were investigated by use of X-ray diffraction (XRD), Scanning Electron Microscopy (SEM) and thin section analysis. After the slant shear test, shards from the mortar surface, repair material surface and biodeposition layer were manually collected using a spatula.

A copper X-ray tube was used for the XRD analysis. The samples were manually trimmed into about 1 cm in diameter and 2 mm to 3 mm thick pieces to fit the sample holder. Scanning was performed from 10° to 70° two-theta with a step size of 0.039°.

Samples for SEM analysis were first subjected to a gold coating process. SEM analysis was performed on an instrument equipped with an EDX detector operating at an acceleration voltage of 20 kV.

To study the formed crystals near the interface between the mortar substrate and the repair material, thin sections (40 mm × 25 mm × 25 µm) were prepared from the tested slant shear specimens, perpendicular to the interface and along the height of the cylindrical specimen (Figure 1). First, the specimens were cut to expose 40 mm × 25 mm faces, which were then glued on a glass slide with a thickness of 2.9 mm. The combined sample was cut and polished until a height of the specimen and glass of 10.1 mm was reached. Next, the specimens were impregnated under vacuum with a fluorescent epoxy. The excess epoxy was polished away and an object glass was glued on the smooth surface. Finally, the glass slides were cut off and the remaining part was polished until a thin section with 25 µm thickness was achieved. A cover glass was glued on top to protect the thin section. The thin sections were then analysed under normal and fluorescent light.

[1] Certain commercial products are identified in this paper to specify the materials used and the procedures employed. In no case does such identification imply endorsement or recommendation by Ghent University or the National Institute of Standards and Technology, nor does it indicate that the products are necessarily the best available for the purpose.

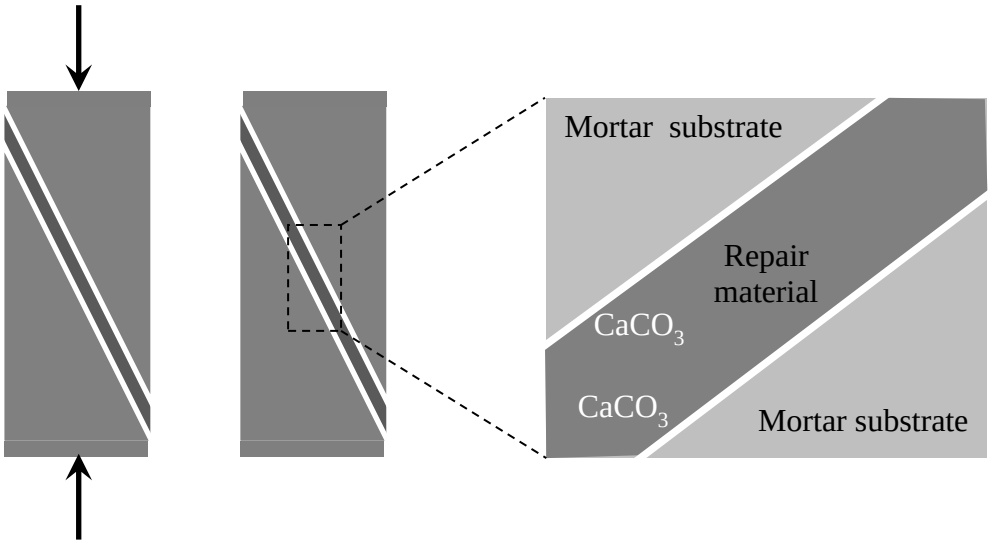


Figure 1: Slant shear setup (left) and thin section location (right)

3. RESULTS

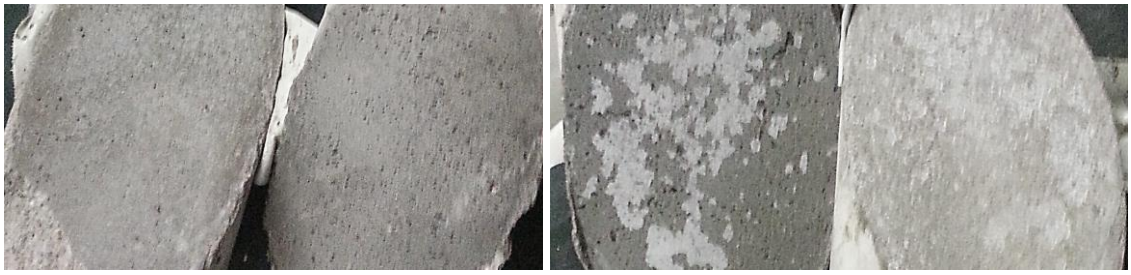
3.1 Slant shear test

The results obtained when performing a compression test on the slant shear specimens, are shown in Table 1. The elliptical surface area was measured after the test. Possible defects at the edges and larger air voids were subtracted from the complete area. No distinct differences in elliptical surfaces were found on the studied samples series and the results thus could be compared. The bond strength results were characterized by an average and standard deviation of 12.6 MPa ± 1.6 MPa and 14.3 MPa ± 1.0 MPa for the REF and BAC specimens, respectively. The average results for the bacterially treated specimens are 13% higher compared to the un-treated specimens. The bacterial treatment thus seems to improve the overall bonding strength, but this preliminary result is only significantly different at a significance level of 0.18 (t-test). The BAC III specimen showed a lower force when performing the slant shear test for unknown reason. Additional future tests will elucidate this assumption and an improved treatment will be proposed.

Table 1: Force, elliptical surface area and bond strength results

	Force [kN]	Elliptical surface area [cm²]	Bond strength [MPa]
REF I	99.15	9120.6	10.9
REF II	117.04	9131.6	12.8
REF III	126.78	9040.3	14.0
BAC I	135.7	9148.5	14.8
BAC II	136.21	9143.8	14.9
BAC III	119.99	9079.6	13.2

Figure 2 shows the elliptical surface after performing the slant shear test. The surface of the reference samples is smooth and debonding of the repair material layer from the mortar surface occurred. Conversely, a clear whitish layer was observed on the bacterially treated specimens. This is from the calcium carbonate biodeposition due to the bacterial treatment. Partial debonding of the calcite layer deposited by the bacteria was seen by comparing the bonding surfaces after compression (Figure 2). This rougher surface is thus possibly primarily responsible for the increase in slant shear bond strength.



REF BAC

Figure 2: Bonding surfaces after performing the slant shear test

3.2 XRD analysis of the carbonates

The XRD spectra are shown in Figure 3 and the EDX spectra in Figure 4. The biodeposition film on the mortar surface mostly consisted of calcite and vaterite. The amount of both was quite significant, which is indicated by the sharp and strong dominant diffraction peaks in the spectrum (green lines). The compositions of the mortar substrate and repair material surfaces were quite similar, as they are both cementitious materials. They both contained calcite, vaterite and quartz. The detailed percentage of each mineral is unknown from this qualitative analysis. Yet it can still be seen that calcite was the main mineral, while the amount of vaterite was much less than that of calcite on the mortar substrate and repair material surfaces. This can be judged by the fact that the dominant diffraction peaks of vaterite were very weak in the spectra of the samples from mortar substrate and repair material surfaces (blue and red lines). No calcium hydroxide (CH) or calcium silicate hydrate gel (C-S-H) was found on the surfaces, suggesting that they were highly carbonated. Nor was aragonite indicated in any of the XRD spectra.

Vaterite is the major product in the pH-range between 8.5 and 10. Conversely, aragonite preferably forms at pH 11, while calcite is the dominant product when the pH is higher than 12 [9]. Vaterite is a metastable polymorph of calcium carbonate and is rare in natural environments [10]. It is unstable and transforms to calcite (or aragonite) at room temperature in an aqueous solution [11]. However, vaterite can be synthesized in chemical processes and often forms in the presence of microorganisms [12]. This gives an indication that bacteria and their secretion (mainly organic compounds) may facilitate the formation of vaterite.

The three kinds of surfaces had completely different morphologies. No obvious crystals were observed on the mortar surface. While on the repair material surface, particles of a size ranging from 2 µm to 5 µm were seen. Based on the EDX spectrum, these particles might be Ca-Mg-carbonates.

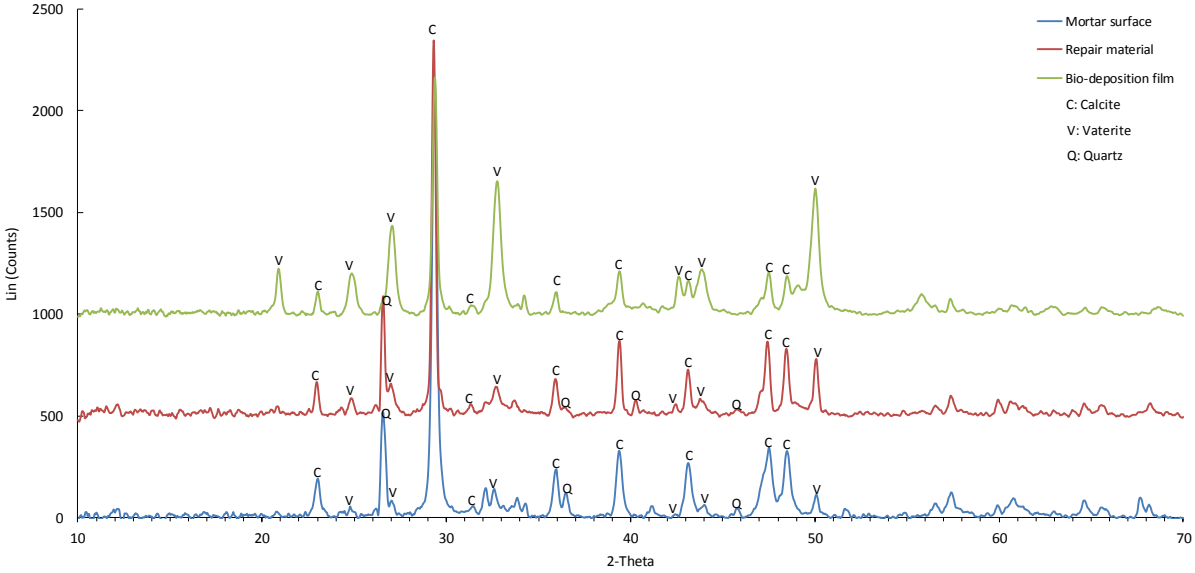


Figure 3: XRD spectra from mortar substrate, repair material and bio-deposition surfaces

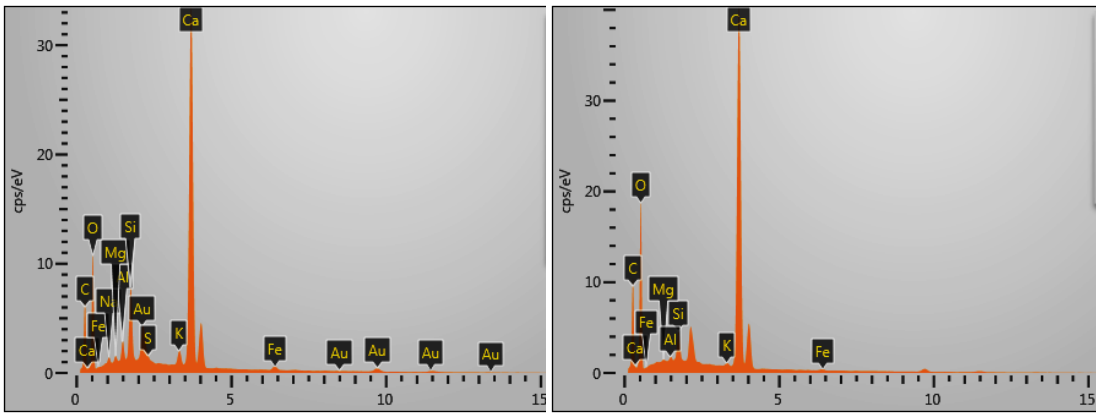


Figure 4: EDX spectra from the mortar substrate (left) and the repair material (right) surfaces

The biodeposition film was full of bacterial imprints. These imprints could be seen as long elliptical shaped spots with a length of approximately 2 µm. The EDX spectra indicated that the film was mainly composed of calcium carbonate. The film was not flat; instead, it was rough with a lot of pits. This could be due to roughening of the mortar surface with sand paper.

3.3 Thin-section analysis

Thin-section analysis is useful in viewing the interlayers between the mortar and the repair product in case of the reference samples and the interlayers between the mortar, calcium carbonate (biodeposition) layer and the repair material, respectively, for the bacterially treated specimens. In the case of the reference sample, the crack propagated through the interface of the mortar substrate and the repair material. This seems to be the weakest link when applying the repair material.

In the case of the bacterial treated specimens, it could be seen that debonding takes place in the repair material near the vicinity of the $CaCO_3$ layer or in the interface between the layer and the substrate. Both the bonding with the mortar substrate and the repair material seems to be sufficient to increase the bond strength.

The complete system in the bacterially treated specimens is shown in Figure 5. A clear whitish layer can be seen in between the mortar substrate and the repair material.

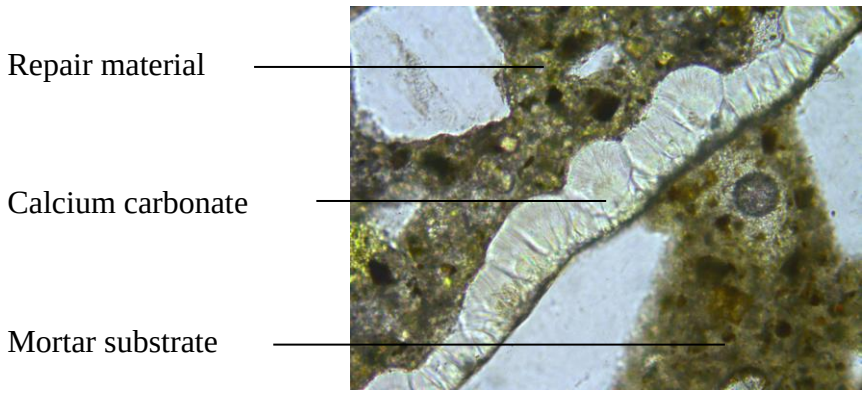


Figure 5: Formed $CaCO_3$ layer attributed to bacterial activity near the interface
The figure width corresponds to 800 µm

The whitish $CaCO_3$ layer in the bacterial specimens has an average thickness of 52 µm ± 14 µm (n = 250). No corresponding white layer can be found in the reference specimens.

The formed $CaCO_3$ layer has a rough surface due to the formation of irregular crystals. Both the repair material and the mortar substrate tend to show a good adherence to that layer. This roughness could also partially lead to the increase in observed bond strength. Future tests will elucidate this assumption.

4. CONCLUSIONS

Based on this preliminary study of applying a biodeposition layer for bond enhancement of repair materials, the following conclusions can be drawn:

- A bacterial treatment has been shown to enhance the bonding of a repair product to a mortar substrate. Further research will focus on how to further optimize the bacterial layer to obtain a significantly higher bonding strength.
- The biodeposition film on the mortar surface mostly consisted of calcite and vaterite.
- Both the mortar substrate and the repair material exhibited a good bonding with the calcium carbonate crystals precipitated by the bacteria.

Ongoing studies are focusing on further enhancing the bond strength of the composite system by further engineering of the biodeposition layer.

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