Study of biocementation as crack sealing technique

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Abstract

The purpose of this work was to study biocementation as crack sealing technique and compare this treatment with one traditionally used for this application (acrylic mastic Sikacryl S). Cracks in stone samples with an opening of 1 mm and with an artificially created rough surface were studied. Permeability tests were carried out to evaluate the sealing of the crack during the treatment by biocementation, which lasted 20 days. Ultrasonic pulse velocity, measured perpendicularly to the gap, was measured after the application of the treatment to evaluate the homogeneity of the treatments and diametral compression tests were carried out to find tensile strength and evaluate the adhesion of the treatments. After carrying out these tests, the contact surfaces with the biocement were analyzed through mineralogical analysis of the precipitate, and by photographs and images obtained by a stereomicroscope. The topography of the biocement precipitated in one of the faces of one crack enabled the verification of the heterogeneity of the treatment. It was possible to observe that the adhesion between the crack surfaces was made at only by few contact points, which explains the lack of sealing, however the tensile strength values in average were 46,28% of the average values measured for the intact stone. The results obtained indicate that the traditional treatment presents better results in all the tests carried out, however biocementation is a promising treatment as the results indicate a strong adhesion between the biocement and the stone.

Keywords: Biocementation, biocement, crack, stone, bacteria, tensile strength.

1. Introduction

There are currently, on the market, several "traditional techniques" effective for repairing cracks. However, these techniques have some inconveniences, for example, in terms of sustainability and durability. In this way, new, more ecological techniques have been developed to make the available solutions for treating cracks more environmentally friendly. Biocementation is one of these “green” techniques and consists in the precipitation of calcium carbonate, using ureolitic non-pathogenic bacteria fed with a solution containing calcium. The occurrence of biocementation inside cracks creates calcium carbonate, which is a form of “cement” that fills the empty spaces and thus enables the recovery of the structures' mechanical capacities.

This is an experimental work whose main objective is to study the feasibility of the biocementation process as an alternative to techniques traditionally used to repair cracks in stone substrates. It is part of the work carried out at IST by Barroso [5].

2. Biocementation

Biocementation is a biological process that consists of using microorganisms to produce calcium carbonate, more specifically, the formation of calcite crystals, called biocement. The microorganisms used are the non-pathogenic bacteria *Sporosarcina Pasteurii*. For this process to occur, a solution (feeding solution) consisting of calcium chloride and urea ($CO(NH_2)_2$) must be added. The reaction starts with the hydrolysis of urea (Eq. 1) which is catalyzed using the urease enzyme present in bacteria.

$$CO(NH_2)_2 + 2H_2O \rightarrow CO_3^{2-} + 2NH_4^+ \qquad (1)$$

This reaction results in the formation of carbonate ions (CO_3^{2-}) and ammonium (NH_4^+).

The formed carbonate ions will react with calcium ions (Ca^{2+}) from the calcium chloride present in the feed medium (Eq. 2).

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3 \qquad (2)$$

The reaction forms calcium carbonate ($CaCO_3$), in the mineral form of vaterite or calcite, which is called biocement.

This technique has already been tested in several situations, such as: repairing cracks in concrete structures [1]; consolidation of porous stones [2]; conservation of monuments [3]; repairing cracks in rock and natural stone [4]. The study performed at IST by Cardoso et al [4] is of special interest because the treatment was applied in cracks of various thicknesses and with smooth and rough surfaces. The treatment was carried out for 13 days by submerging the samples in bacteria and feeding solution. The volume of water flow through the crack was measured every 2 days to evaluate sealing achieved by accumulated precipitation of biocement into the crack surfaces. Complete sealing was achieved in almost all the cracks. Indirect tensile tests performed by diametral compression were performed at the end of the treatment and showed that the tensile strength increases with the increment of track width, both for the cracks with smooth surface or rough surfaces. In general, biocementation is more efficient in thicker cracks (1 mm) with a rough surface. The results of this study showed the efficiency of the treatment and inspired this work.

3. Materials and experimental setup

3.1. Stone samples

The stone tested was cut from borehole samples of basalt. Minerals present are magnesium, aluminum, silicon, potassium, calcium, titanium and iron. The dry unit weight is 27,26 kN/m^3 and porosity is 0,79%.

A 72 mm diameter core from this rock was cut into 9 stone disks approximately with thickness of 38 mm, in the zones appearing not to have discontinuities. From these, 3 (I1 to I3) did not need further preparation as they were used as a standard and comparison term for the treatments performed. The other 6 disks were cut, using a saw, into two pieces along their diameter, to create artificial discontinuities with smooth surface. The 12 faces of the cuts were submitted to a process that intended to simulate roughness on the crack surfaces. Such roughness was obtained using a hand saw, creating cracks, with a depth of between 0,5 and 1,0 mm and a distance between them of 8,72 mm, in the form of a mesh (Figure 1).

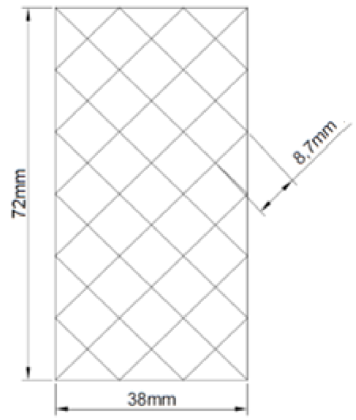

Figure 1 - Mesh-shaped pattern on the surfaces in contact with the treatments.

The two halves were assembled using plastic spacers with a thickness of 1 mm, creating a joint along the diameter of the disk, with constant width. The lateral cracks of the disk were wrap with a tape and the disks were wrap with a nylon clamp to keep the joint geometry. In total, 6 disks were made from which 3 were used for testing the biocement treatment (B1 to B3) and the remaining 3 were used for testing the traditional treatment (T1 to T3).

3.2. Treatments

The bacteria *Sporosarcina Pasteurii* grown in culture medium (20 g/L yeast extract, 10 g/L of ammonium sulphate and 0,13 M Tris buffer pH 9.0) at 30°C in an incubator was used for the treatment by biocementation. Bacteria grew to a cell density of ~10^8 cells/mL, corresponding to an optical density of 1 measured for 600 nm (OD_{600}).

The feeding solution was prepared with 0,5 M of urea, 0,5 M of calcium chloride (calcium source), 1:10 diluted growth medium, 2,12 g/L of sodium bicarbonate and 10 g/L of ammonium chloride.

The product used as a traditional treatment is Sikacryl S, a one-component plastic mastic, based on an acrylic dispersion. It is a non-drip sealant, normally used in cracks with little movement (allows movements up to 7.5% of the crack width) in materials such as concrete, mortar, brick, aluminum, PVC, wood, among others. Can be applied to cracks if they are not permanently immersed in water.

3.3. Application of the treatments

The biocementation treatment was applied using a system created solely for this purpose (Figure 2). The system consists of a PVC container (so that it does not react with the bacteria feeding solution) divided in two cells that can be separated, so it is possible to access both the upper and the lower face of the sample. The stone disc was placed in a circular opening, with a diameter slightly larger than the diameter of the disc, being the gap left sealed with silicone to prevent leakage of the feeding solution into the lower cell. Before placing the disc in the system, it was necessary to create a watertight reservoir corresponding to the volume of the crack, that is, to prevent bacteria from flowing through the crack into the lower cell at the time of application and, in this way, minimize the volume of bacteria used. The reservoir corresponding to the crack volume was obtained by covering the inferior face of the disk with adhesive tape and then covering the entire inferior face with molten paraffin and waiting for it to solidify.

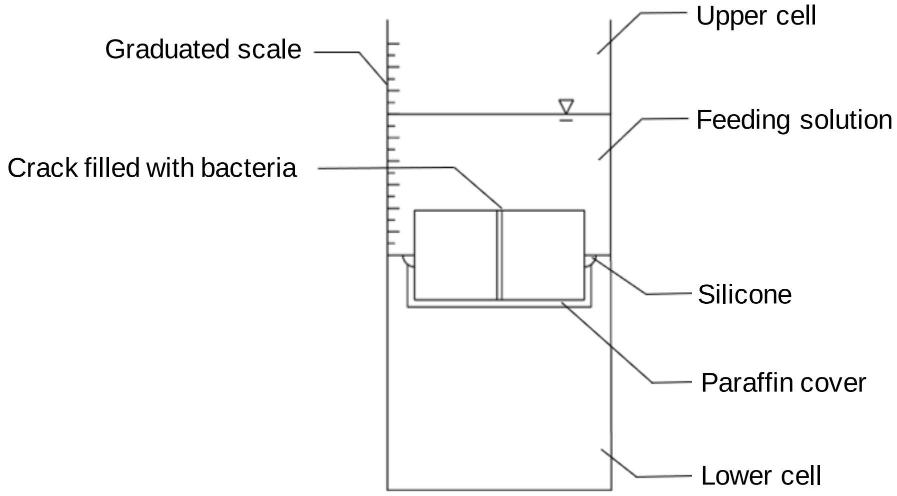

Figure 2 - Biocementation treatment application system.

The duration of the treatment was 20 days. Bacteria were added in 2 stages, each stage lasting for 3 days. The first phase started on day 0 and the second on day 6. During the 3 days of application of the bacteria, the bacteria solution was injected into the crack and then the disc was submerged in feeding solution for one day without the bacteria being purged. On the remaining days, between the 3rd and 6th and from the 6th to the 15th, the sample was submerged in feeding solution, with the submersion being interrupted every 3 days to carry out a test to obtain the permeability of the cracks, using water. This test was also done before starting the treatment as well. To complete the biocementation treatment, the disc was kept submerged in feeding solution for the last 5 days without neither purging nor adding further bacteria and feeding solution.

The application of the traditional treatment (mastic) on the crack was done using a spatula with a thickness of 0,2 mm as the mastic high viscosity prevents it to flow. The spatula allowed the treatment to be better distributed and to present better homogeneity along the two faces of the crack. The crack was filled with the mastic.

3.4. Tests performed

Tests were carried out to assess the quality of the treatments. These tests were:

- Volume of water flowing through the cracks, to evaluate the efficiency of the treatments in their behavior as sealants. Permeability was measured using the permeability coefficient (K), obtained from the water flow speed.
- Ultrasonic pulse velocity, measured perpendicularly to the gap, evaluated the homogeneity of the treatments. Exponential transducers were used, instead of the traditional cylindrical transducers, because the lateral faces of the disks are circular, and these transducers allow a better contact between the lateral face of the disks and the faces of the transducers.
- Diametral compression tests were carried out to find tensile strength and evaluate the adhesion of the treatments. The samples were placed on the loading platform so that the cracks were perfectly vertically aligned with the load cell.

After performing the tensile strength tests by diametrical compression, the discs were separated by the cracks to make a more accurate assessment of the treatment that took place inside the cracks. The analysis performed were as follows:

- Mineralogical analysis by X-ray Diffraction Analysis, XRD, to characterize the mineral form of calcium carbonate precipitated and by Scanning electron microscope images, SEM, to see if the minerals appear to be calcite (rhombohedra shape) or vaterite (spherical shape).
- Image analysis to understand the homogeneity and deposition patterns of the precipitated biocement. The faces of the stone in contact with the precipitated material were photographed at the macro level and using a stereomicroscope.
- Topography of the biocement covering the crack to evaluate the crack filling volume. It was used an application named “eyesCloud3d” which allows the generation of 3D models, through 5 photos taken with the mobile phone.

4. Results

4.1. Permeability

Figure 3 shows the progress of the permeability coefficient (K) over the 20 days of treatment by biocementation. As expected, in all samples this coefficient was always decreasing throughout the treatment because the cracks were becoming filled with the precipitated biocement. However, it was not possible to reach no flow at all, indicating that the cracks were not completely sealed. The treatment was interrupted but it would probably have been possible to further decrease the permeability (and fill the cracks more) because the permeability rate was still decreasing. Sample B1 had the lowest permeability coefficient among all samples, with a value of 3.50E-02 m/s and, therefore, this was the sample in which the best sealing was achieved.

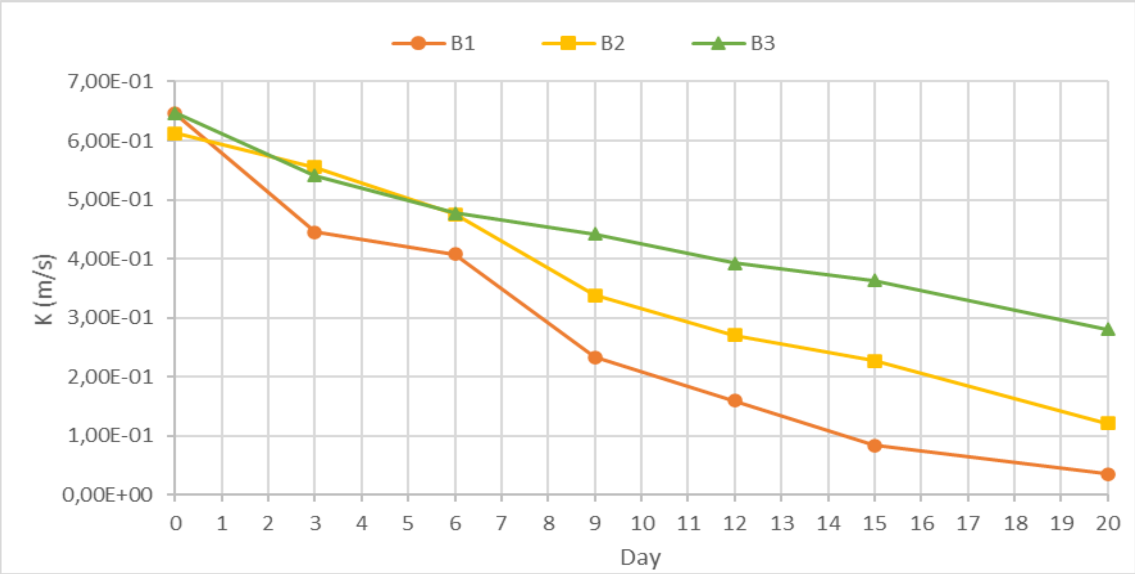

Figure 3 - Curves of the evolution of the permeability coefficient (K), per sample, throughout the treatment.

At the end of the application of the traditional treatment, an attempt was made to determine the permeability coefficient of samples T1 to T3, but this was not possible because there was no water running through the crack. This indicates that these samples were completely sealed.

4.2. Ultrasonic pulse velocity

Figure 4 shows the average values of the ultrasonic pulse velocity measured in the intact samples and in those with traditional treatment and treated by biocementation.

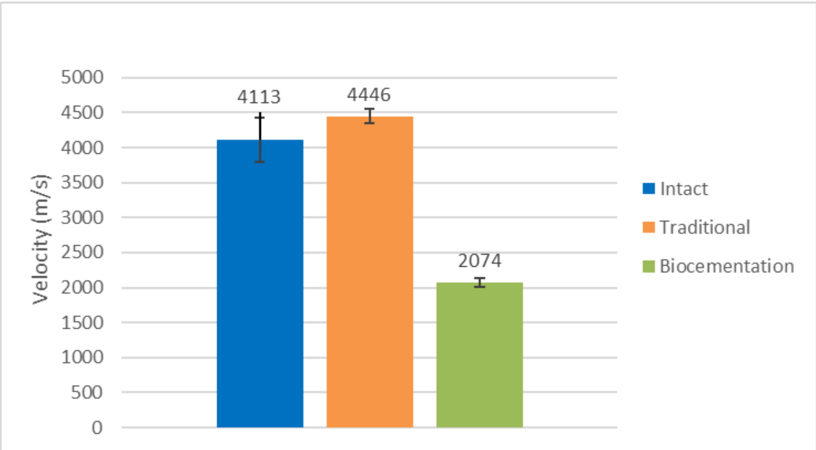

Figure 4 - Average values of ultrasonic pulse velocity readings.

The traditional treatment presents an average velocity of ultrasonic pulse larger than that of the biocementation treatment. Compared to the biocementation treatment, the traditional treatment provides a more homogeneous filling of the crack. The average of readings for intact samples is lower than for traditional samples. This is due to the presence of discontinuities in the intact samples, which were not considered when choosing the specimens.

4.3. Diametral compression tests

The average values of tensile strength of intact samples, with traditional treatment and treated by biocementation can be seen in Figure 5.

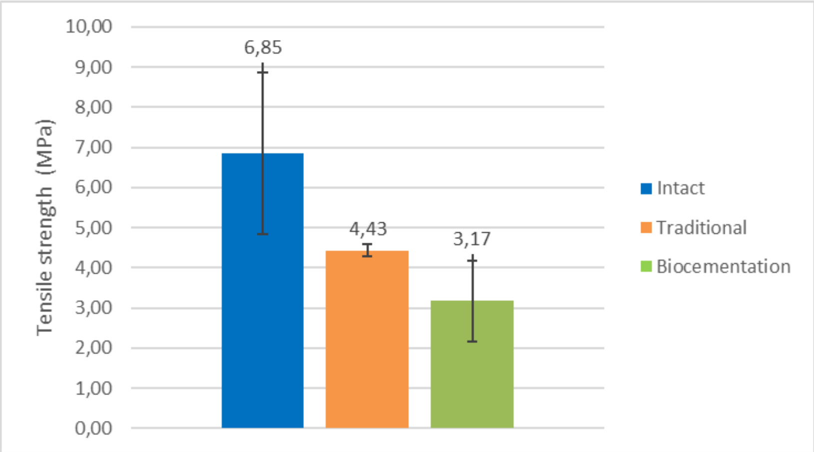

Figure 5 - Average values of tensile strength by diametrical compression.

Comparing the average values of the tensile strength measured by diametrical compression of the samples treated traditionally and samples treated by biocementation with the values of the intact samples, it is possible to verify that the samples treated by the traditional method have values closer to the values of the intact samples.

The relation between the values of the tensile strength measured on the treated samples and that of intact samples, in percentage, is presented in Table 1 for both treatments. Based on these values it can be concluded that the adhesion achieved for the traditional treatment is better than that for the biocementation treatment.

Table 1 - Comparison of the tensile strength obtained after each treatment in relation to the intact sample (average values).

	Tensile strength after treatment / Tensile strength intact sample (%)
Traditional	64,60%
Biocementation	46,28%

4.4. Mineralogical analysis

The highest “Mineral Score” values resulting from the X-ray diffraction analysis of the samples tested with biocementation treatment and the upper face of sample 1 (FS1) are presented in Table 2. The higher these values, the greater the amount of this mineral in the analyzed sample. From these values it is possible to notice the presence of vaterite and calcite, both calcium carbonate minerals. This result indicates that biocementation had occurred. Vaterite values are generally higher than calcite values in all samples.

Table 2 - Mineral Score values of the minerals present in the samples.

Mineral Score	B1	B2	B3	FS1
Calcite	8	58	10	25
Vaterite	84	63	70	51
Labradorite	-	-	-	20

The presence of calcium carbonate minerals was also detected in SEM images of the precipitate cover removed from the crack (Figure 6). It was possible to observe the precipitation of spherical form of calcium carbonate (vaterite) (Figure 6 (a)). It is also possible to observe marks of places where the bacteria became lodged (imprints), represented with red arrows in Figure 6 (b).

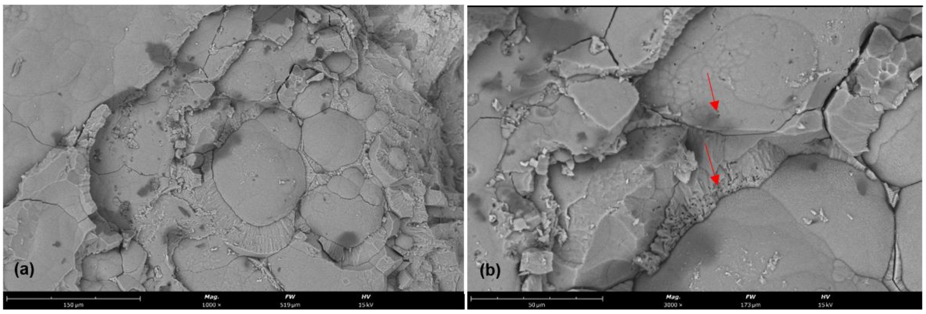

Figure 6 - SEM image of formed crystals (a) and bacterial activity (b) in the crack.

4.5. Image analysis

By analyzing the macro photographs taken from the contact surfaces of each of the samples (Figure 7), sample B1 (Figure 7 (a)) presents a more homogeneous filling of the crack face and sample B3 (Figure 7 (c)) presents the least homogeneous filling.

(a) (b) (c)

Figure 7 - Contact surfaces of samples B1 (a), B2 (b) and B3 (c) after treatment.

Even though the crack was filled with biocement, the grooves that simulate roughness are still visible, even if they are covered with this material. A complete sealing of the crack did not occur and, therefore, the water used in the permeability test continued to flow until the end of the treatment.

4.6. Topography of the precipitated biocement layer

The topographic profile obtained through the mobile application is shown in Figure 8. The existence of peaks confirms that there was a physical connection between the two faces of the crack, but it was punctual. For this reason, tensile strength was measured but complete sealing was not achieved.

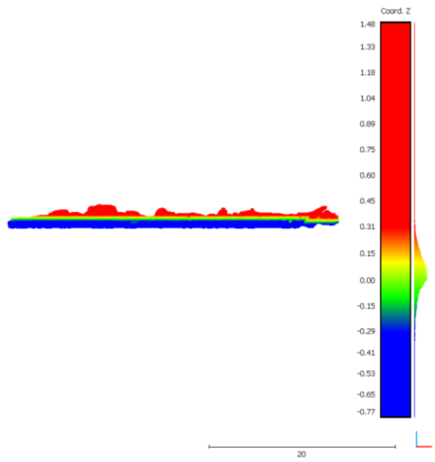

Figure 8 – Topographic profile of the crack biocement cover

5. Conclusions

In this work, the efficiency of two types of treatment for the sealing of cracks was investigated, namely, a traditional treatment, which consists in the application of an acrylic mastic in the crack, and an innovative treatment, the treatment by biocementation.

From the permeability tests it is possible to conclude that the biocementation treatment did not allow the cracks to be completely sealed, at least in the 20-day treatment application time. This is explained by the fact that the artificially created grooves that simulate roughness were not completely sealed, as these created a system of channels through which the solution circulated.

Through the ultrasonic pulse speed tests, it is verified that this speed is higher in samples treated by the traditional method than when biocementation is used. In general, the traditional treatment presents a more homogeneous sealing of the crack, making it completely watertight.

The analysis of the tensile strength, it was found that the percentage variation between the values of tensile strength of intact samples and those of samples treated traditionally and by biocementation is, respectively, 64.60% and 46.28%. Compared to samples treated by biocementation, samples treated by the traditional method have greater tensile strength.

From the mineralogical analysis, which is the conjugation of information resulting from the analysis of X-ray diffraction (XRD) and the analysis with a scanning electron microscope (SEM), it was found that the samples treated by biocementation had carbonate calcium crystals, such as calcite and vaterite. These results allow us to conclude that, in fact, biocementation occurred, even if this did not happen in the same way in all samples.

With the analysis of photographs, it was possible to verify that sample number 3 had a less homogeneous coverage, when compared to samples numbers 1 and 2. This analysis came to prove what had already been observed in the results obtained in the mineralogical analysis.

Through the topography of the crack coverage of sample number 1, it was verified that the connection between the faces is punctual, and it was concluded that the crack was not filled with biocement. As such, a complete sealing of the gap has not been achieved.

In general, after analyzing all the results of all the tests performed, it can be concluded that the traditional methods for treating cracks present better results than the treatment by biocementation. Despite this, biocementation is an innovative and promising method, which with some developments, soon, will become a sustainable and viable alternative for use in construction.

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