Proceedings of the 4th World Congress on Civil, Structural, and Environmental Engineering (CSEE'19)
Rome, Italy – April, 2019
Paper No. ICGRE 143
DOI: 10.11159/icgre19.143

Efficiency of biocementation as rock joints sealing technique evaluated through permeability changes

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Abstract **-** In Microbially Induced Carbonate Precipitation (MICP), bacteria are used to hydrolyze urea. In the presence of a calcium source supplied in a feeding solution, calcium carbonate is formed and precipitates. MICP has shown promising results in terms of improving the hydro-mechanical properties of sandy soils by forming bonds connecting the particles. Recent studies are focused on using MICP for sealing discontinuities, such as concrete and stone cracks, and rock joints. This is investigated in this paper for a disk-shaped rock sample having a crack along the entire diameter. In the study presented, enzyme is used instead of bacteria, because prior studies proved that production of calcium carbonate is faster while using enzyme. In addition, large quantities of enzyme required for Civil Engineering applications can be produced easier comparing to bacteria, and for this reason, using enzyme may be an alternative to using bacteria. The efficiency of the method was evaluated by constant head water permeability test during the treatment. The permeability of the crack reduced along time and the crack was almost completely sealed after 6 hours of treatment. Upon completion of the treatment, the crack was investigated visually to detect the presence of precipitated biocement.

Keywords**:** Biocement, Enzyme, Discontinuity, Sealing, Rock joint, Permeability.

1. Introduction

Natural stone is being used as construction material and foundation materials. Discontinuities at rock such as cracks and joints are the main cause of the instability and deterioration, in particular in historical structures (i.e. ancient cathedrals, historical buildings, monuments and statues), as well as in underground construction within the rock masses (i.e. tunnels and other underground excavations). So far, several treatment methods have been developed to heal these flaws, each with their own disadvantages (material incompatibility, irreversibility).

Microbially Induced Carbonate Precipitation (MICP) has been developed recently in the field of geotechnics as an environment friendly technique. In MICP, bacteria enzyme urease catalyzes the hydrolysis of urea (Eq. 1). After a series of acid-base reactions (Eq. 2 and 3) carbonate CO_3^{2-} is formed, as well as ammonia NH^{4+} (Eq. 4) [1]. If this process takes place in a calcium-rich medium, then calcium carbonate $CaCO_3$ is formed (Eq. 5). This biocement is usually called as calcite.

$$CO(NH_2)_2 + H_2O \rightarrow NH_2COOH + NH_3 \quad (1)$$

$$NH_2COOH + H_2O \rightarrow NH_3 + H_2CO_3 \quad (2)$$

$$H_2CO_3 \rightarrow 2H^+ + 2CO_3^{2-} \quad (3)$$

$$NH_3 + H_2O \rightarrow NH^{4+} + OH^- \quad (4)$$

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3 \ (K_{SP} = 3.8 \times 10^{-9}) \quad (5)$$

MICP has shown promising results for improving hydro-mechanical properties of sandy soils via production of biocement as bonding agent. Satisfying results have been reported in different application of MICP i.e. reducing the liquefaction potential of sands [2] increasing the shear strength and decreasing permeability of soil [3], [4] and [5] surface crack remediation and improving durability of concrete [6], enhancing the strength and water absorption of cement-sand mortar [7], improving the compressive strength and permeability of shotcrete [8]. Recently more researches are developed concerning the application of MICP as a sealing technique for cracks. Choi et al. [9] studied the relation between opening of a crack and the efficiency of MICP treatment on several mortar samples. Tittelboom et al. [10] compared the crack healing potential of bacteria and traditional repair technique on concrete samples by means of water permeability, ultra sound transmission measurement and visual examination. They stated that bacteria protected in silica gel could induce precipitation of calcium carbonate and fill the crack [10]. Though some researches have been developed about protection of stone using biocementation or biodeposition [11], [12] and [13] but as far as the authors know there are no publications reported about treatment of rock joint using biocementation.

In the present study, a disc-shaped rock sample (61 mm diameter, 30 mm thickness) was cut with saw to have an artificial crack along the diameter. Then it was treated to be sealed with biocement. In most of the similar studies, the sample is soaked into the treatment solution for certain period of time, however in the present study, the treatment solution was flowing through the crack along the treatment procedure. This method may reflects more realistic aspect of the treatment in large-scaled applications. The evolution of permeability during the treatment was used as monitoring tool of its efficiency. After the treatment the crack was open to visually observe the amount and pattern of precipitated calcium carbonate.

2. Materials and methods

2.1. Rock sample

The sample was extracted from core samples 61 mm diameter collected during exploratory works for underground construction in the city of Lisbon. According to the geological report, the rock is grayish Basalt from early Cretaceous, classified as C4B, moderately weathered (W_{3-2}). The rock is fractured with joint spacing between 20 to 60 cm (F_{4-3}) and fractures are filled with calcite (white color spots highlighted in figure 1). RQD of rock mass is reported around 75%. The dry unit weight, γ_d, specific gravity, G_s, and porosity n of the rock were measured and are 26.78 kN/m^3, 2.84 and 3.69% respectively. Figure 1 shows the core from which the disk-shape sample was extracted.

Fig. 1: Core sample taken from underground work in Lisbon.

The core was cut into disks 30 mm thick. One disk appearing not to have discontinuities was cut along its diameter into two pieces, using saw. An artificial crack was created by attaching the two pieces to each other using installation tape. The irregularities on the crack surfaces caused by saw left the crack open. The opening of the crack was measured using microscope along the crack and for both sides of the sample. The average opening was 0.93 mm at the top-end and 0.31 mm at the bottom-end. These values are typical crack width in such type of rock.

2.2. Treatment solution

2.2.1 Enzyme

An enzyme solution (urease from *Canavalia ensiformis*, Jack bean, supplied by SIGMA) with initial concentration of 15 g/L was prepared with distilled water in the laboratory and stored in -20˚C until the treatment started. The final concentration of enzyme in treatment solution was 1.5 g/L. From past studies it is known that calcium carbonate resulting from activity of this enzyme is in the form of calcite, which is a stable insoluble form of calcium carbonate [14].

2.2.2. Feeding solution

The recipe for calcium-rich feeding solution is presented in table 1. The main components of the feeding solution are the urea and calcium chloride, which is the source of calcium. The other components of the feeding solution are basically to enhance the pH of the medium and to feed bacteria (if used). Although enzyme is the catalyzer, it was decided to use similar feeding solution for future comparison when bacteria is used. Feeding solution was stored at 4˚C.

Table 1: Cooking recipe for feeding solution.

Component	Concentration (g/L)
Urea $CO(NH_2)_2$	30.04
$CaCl_2$+H_2O	73.50
$NaHCO_3$	2.12
NH_4Cl	10.00
$(NH_4)_2SO_4$	1.00
Yeast Extract	2.00

2.2.3. Experimental setup and treatment

In this research, the idea was to keep the treatment solution flowing through the crack instead of submerging the sample into it. Figure 2, presents the experimental setup.

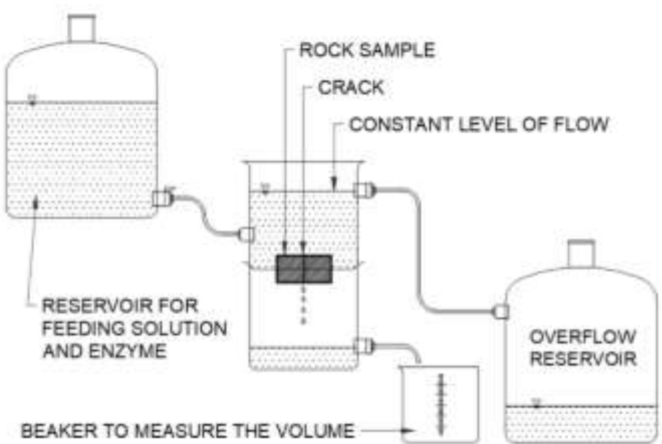

Fig. 2: Set up for treatment and permeability test

The surface of rock sample was covered by tape except for a few millimeters wide strip along the crack, in order to avoid penetration of solution into the sample from anywhere other than the crack. Then, the sample was fixed between to containers and sealed, so the solution would pass just through the crack. The enzyme and feeding solution were mixed and immediately loaded to the setup and by opening the valve, solution flew over the sample and started passing through the crack. The head of solution above the crack was kept constant (at 6 cm above the sample) by means of collecting the overflow. The solution collected at the overflow reservoir and measurement beaker was reloaded to the main reservoir.

2.3. Evaluation

2.3.1. Permeability

As treatment was applying, at certain time intervals, the volume of the solution collected at measurement beaker was recorded as a representative of permeability of the crack. The flow was calculated using equation 6,

$$\boldsymbol{Q}\left(\frac{\boldsymbol{L}}{\boldsymbol{s}}\right)=\frac{\boldsymbol{V}\ (\boldsymbol{L})}{\boldsymbol{t}\ (\boldsymbol{s})} \tag{6}$$

where (Q) is the flow passing the crack and (V) is the volume of the solution collected at measurement beaker at time duration of (t).

2.3.2. Visual inspection

After the treatment was completed, the setup was dissembled and the rock sample was dried out in laboratory temperature and then the crack was detached using a cutter. This allowed to have the two surfaces of crack and visually investigate them to observe the amount and pattern of precipitated $CaCO_3$.

Calcium carbonate is not soluble in water when it is in the form of calcite [15] while the components of the treatment solution (enzyme, urea and calcium chloride) are. Therefore, in order to confirm that the precipitated crystals are in the form of calcite, a simple solubility test was performed by adding few drops of water on the crystals and evaluate their dissolution. The XRD analysis (X-Ray Diffraction) could not be performed because the amount of precipitation was not enough for such test.

3. Results and discussion

3.1. Permeability

The flow passing the crack was measured during the treatment as representative of permeability of the crack. Figure 3 (a) presents the flow passing through the crack versus time, computed using eq. 6.

The volume of solution passing through the crack was reduced along the time from 3.3×10^{-3} m^3/s to 2.0×10^{-4} m^3/s only within almost 5 hours of treatment, which proves the efficiency of treatment. The calcium carbonate was formed and precipitated in the crack and sealed it. For the first hour of treatment, the flow reduced from 3.3×10^{-3} to 3.1×10^{-3} (only 6.06% reduction) but for the period between 1 to 3 hour of treatment (step 1), the flow dropped from 3.1×10^{-3} to 0.9×10^{-3} (70.96% reduction). The rate of reduction again reduced between 3 and 4.25 hour of treatment (11.11%) and one more time (step 2) the flow was decreased from 8×10^{-4} to 2×10^{-4} (75% in 1 hour). Figure 3 (b) presents the rate of reduction in flow versus time, highlighting the two steps mentioned.

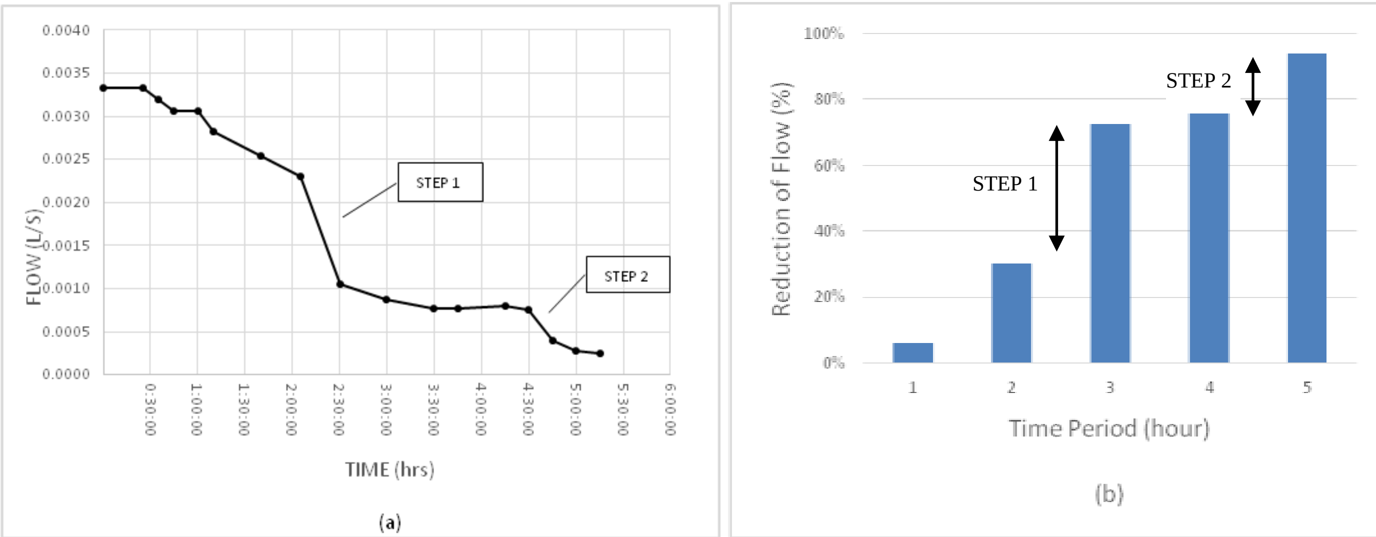

Fig. 3: (a) Flow passing through the crack versus time. (b) Reduction of flow during the treatment versus time.

3.2. Visual inspection

Figure 4 shows a photograph of the detached crack, where it can be seen the calcite crystals (from original texture of rock) and a precipitated material. This material is distinct from natural calcite minerals (both white color) due to texture. The crack surface was flat after saw and become rough after the experiment. The test carried out by pouring few drops of water on the precipitated white mineral indicated it can be calcium carbonate because no dissolution occurred.

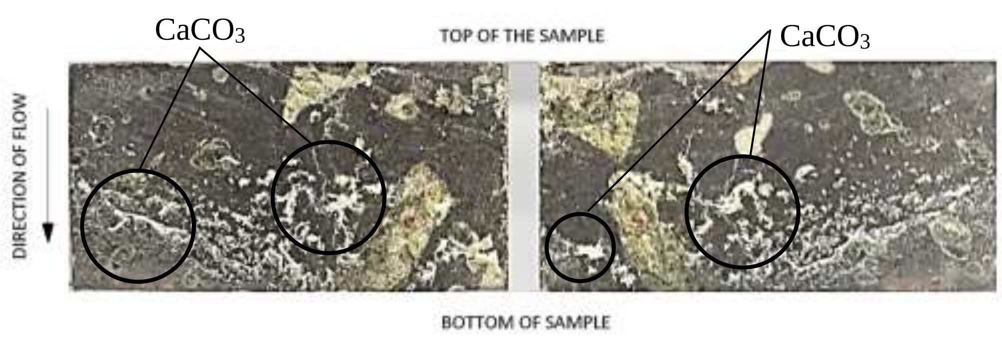

Fig. 4: Precipitated calcium carbonate on the crack surfaces.

As it can be seen in Figure 4, the precipitation has mostly occurred near the bottom side of the sample and no biocement was seen at the top side. This may be explained by the smaller crack openings at bottom sides of the sample. Assuming that the opening of the crack changes on linear basis from one end to another, being larger at the top and smaller at the bottom, this trapezoidal shape favored sealing starting from the bottom.

From data presented it might be concluded that the precipitation of calcium carbonate occurred, sealing the part of crack having opening less than 0.62 mm. This value is within the ranges of crack openings typical for such kind of materials, validating the viability of biocementation for such application.

4. Conclusion

A disc-shape rock sample having a crack along the diameter was subjected to treatment with biocementation. Enzyme was used to induce the hydrolysis of urea and precipitation of calcium carbonate. The setup was built in such a way that the treatment solution flows through the crack throughout the treatment. The crack was sealed after five hours of treatment. Later, the crack was visually inspected and calcium carbonate was observed at places where the crack opening was smaller, proving that biocementation had occurred. The opening of the crack was within realistic values therefore encourage more comprehensive investigation on this treatment for such application. Further researches are planned to study the application of bacteria for biocementation in rock joints. In addition, different geometrical parameters of crack i.e. opening and roughness of the surfaces are to be studied.

Acknowledgement

Financial support of FCT (Fundação para a Ciência e a Tecnologia) through project BIOSOIL, ref. PTDC/ECI-EGC/32590/2017, and cooperation of laboratory of Department of Bioengineering at Instituto Superior Técnico, University of Lisbon, are highly appreciated.

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