

Modeling of Biocalcification in Non-saturated Conditions

Catherine Raymond-Poirier¹ and Benoît Courcelles*¹

¹Polytechnique Montreal

*Corresponding author: C.P. 6079, succ. Centre-Ville, Montréal, Qc, H3C 3A7, Canada,

benoit.courcelles@polymtl.ca

Abstract: Biocalcification is an emerging technique of soil improvement based on the metabolic activity of *Sporosarcina pasteurii*, which consists of the injection of bacteria followed by nutrients, salts and urea in soils. The enzymatic activity results in the precipitation of calcium carbonates providing a cohesion to granular soils and finally improving their mechanical properties. The paper proposes a numerical model implemented prior to laboratory tests to verify the applicability of this method to non-saturated conditions. The results present a good homogeneity of treatment and confirmed its applicability to non-saturated conditions such as river banks or road foundations.

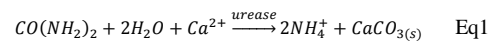
Keywords: biocalcification, non-saturated condition, porous medium, calcite.

1. Introduction

Soil improvement techniques constitute viable alternatives to expensive foundations where superficial foundations can't be implemented. Among the great diversity of soil improvement techniques, the biocalcification of granular soils appears as a new, environmentally friendly and energy-efficient technique. This promising alternative relies on the formation of calcium carbonates based on the metabolic activity of *Sporosarcina pasteurii*. Indeed, the process consists of the formation of precipitates that create a cohesion and finally improve the behavior of granular materials.

The treatment of soils is divided into three steps: (1) injection of bacteria, (2) injection of nutrients, salts and urea, and finally (3) flushing of byproducts. The formation of precipitates occurs after the injection of nutrients, salts and urea through the enzymatic activity of *Sporosarcina pasteurii*. The urea ($\text{CO}(\text{NH}_2)_2$) is consumed within the bacteria and creates ammonia (NH_3) and carbon dioxide (CO_2). After diffusion through the cell wall, two reactions spontaneously occur in the surrounding solution: the ammonia is converted to ammonium (NH_4^+) and the carbon dioxide reacts with the carbonate

system. Consequently to the production of NH_4^+ , hydroxyl ions (OH^-) are generated and lead to an increase of pH. The overall process leads to the saturation of the solution and the precipitation of calcite (CaCO_3) in the presence of Ca^{2+} ions in excess. This excess is obtained through an injection of calcium chloride (CaCl_2). Finally, the precipitates create some bridges between the soil particles and influence the geomechanical and hydraulic parameters such as stiffness, shear strength, hydraulic conductivity, etc. (Dejong et al., 2010). The overall reaction is provided in Eq1.



This technique was initially developed in saturated conditions, but Cheng et al. (2013) proposed a new injection method relying on surface percolation in non-saturated soils. As this new methodology is very promising for slope stability, river banks treatments or road foundations, the present paper aims to verify the homogeneity of treatment prior to a series of laboratory tests. Indeed, a new drip-fed irrigation system that can treat up to 6 samples simultaneously has been imagined at our laboratory and the simulations aim to check its theoretical efficiency to improve its conception.

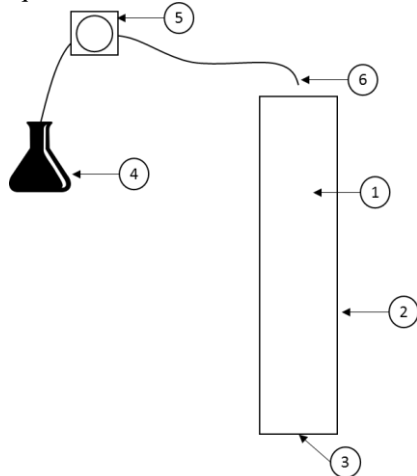
2. Laboratory tests

The laboratory tests consist in treating a column of sand in non-saturated conditions. The samples consist of a cylinder (2 in. in diameter and 4 in. high) treated from the top through a drip-fed irrigation system (see. Fig. 1). The cylinder is filled with a fine sand with grain diameters between 0.08 and 0.55 mm. The grain-size curve is very uniform with C_u and C_c respectively equal to 0.95 and 2.10. The sand is lightly compacted prior to laboratory tests, which results in a porosity of about 0.34.

To ensure a uniform repartition in the system, the bacteria are mixed with the sand prior to its compaction. Then, the reactants (nutrients, urea and calcium chloride) are injected from the top at a flow rate of 3 ml/min thanks to a peristaltic

pump. The drops falling from the peristaltic pump lead to the creation of a small pond in the middle of the top surface of each sample. The method of treatment is summarized on Figure 1.

After treatment, the purpose of the tests is to evaluate the behavior of soils with respect to freeze–thaw events, which are very important considering the potential application of this technique to road foundations.



- 1: Sand and bacteria
- 2: Impermeable cylinder
- 3: Exit (screen)
- 4: Solution (nutrients, urea and calcium salt)
- 5: Peristaltic pump
- 6: Drip-fed irrigation system (up to 6 samples can be connected simultaneously)

Figure 1. Illustration of laboratory tests

2. Modeling

2.1 Hydraulic model in non-saturated conditions

As regards the infiltration of water, the model mainly relies on the Richard's equation for infiltration in non-saturated granular media (Eq2).

$$(C + S_e S) \frac{\partial H_p}{\partial t} + \nabla \cdot (-K \nabla (H_p + z)) = 0 \quad \text{Eq2}$$

where C represents the specific moisture capacity (m^{-1}); S_e the effective saturation (-); S the storage coefficient (m^{-1}); H_p the pressure head (m); K the hydraulic conductivity (m/s) and z the vertical elevation (m).

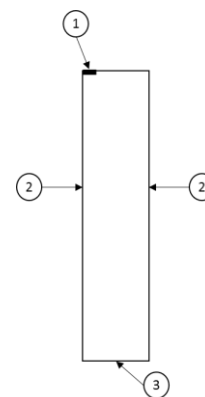
In this nonlinear equation, the retention and permeability properties are evaluated according to the Van Genuchten model. Furthermore, the porosity is updated with respect to the volumetric ratio of (calcite / pore volume) as a result of the formation of precipitates.

Geometry

The model was performed in rotational symmetry and the samples were represented as a cylinder of 2'' in diameter and 4'' high. The percolation was performed from the center of the top surface and the pond created by the fall of drops was represented as a constant hydraulic charge in a ring (5 mm in diameter).

Boundary conditions

As the level of water in the ring was constant and equal to 5 mm, the infiltration of water in the sample was represented as a Dirichlet constraint on pressure head. A no flow condition was applied to the vertical wall as the soil samples are placed into an impermeable cylinder. As the water was drained from the bottom of the samples, an outflow condition was applied to their base. Finally, a no-flow condition was applied to the axis of revolution as a result of the symmetry. The geometry and the boundary conditions are summarized on Figure 2.



- 1: Fixed hydraulic head
- 2: No-flow boundary condition (Rotational symmetry axis)
- 3: Outflow boundary condition

Figure 2. Geometry and hydraulic boundary conditions (no scale)

2.2 Chemical model

This model involves the advection–diffusion–reaction differential equation in saturated porous media and considers a time-dependent porosity as a function of the precipitation of calcite. In our model, the following hypothesis are assumed:

1. Only dissolved species can react,
2. Chemical reactions involved in the process are only hydrolysis of urea and precipitation of calcite,
3. Ca^{2+} ions are considered in excess and all carbonates ions are converted to calcite after hydrolysis (result of the microbial activity),
4. Calcite carbonates are considered immobile after precipitation.
5. No adsorption or surface reaction are considered in the model.

The water in the injection ring contains Ca^{2+} ions at a constant concentration C_0 . These ions enter the sample under the surface ring and move through the sample by advection and dispersion/diffusion. The equation that governs the transport involves advection, dispersion/diffusion and reaction of solutes as given in Eq3.

$$\frac{\partial}{\partial t}(\theta c) + u \cdot \nabla c + \nabla \cdot [-\theta D_L \cdot \nabla c] = \sum R_L \quad \text{Eq3}$$

where c represents the dissolved concentration (mol/m^3), θ the volume fluid fraction (-), D_L the hydrodynamic dispersion tensor (m^2/d) and R_L the reactions ($\text{mol}/(\text{m}^3 \cdot \text{d})$).

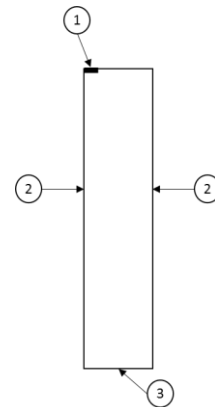
The reaction rate of ureolysis depends on several factors such as the number of bacteria, their growth and storage conditions, as well as environmental parameters such as temperature and pH (Bachmeier et al. 2002, Stocks-Fischer et al. 1999, Whiffin 2004). Moreover, the formation of calcite creates a diffusion barrier around the bacteria that decreases their activity (Bang et al. 2001) or creates anaerobic conditions leading to their death (Whiffin et al. 2007). As a result of these phenomena, a decay of the reaction rate (r) has been assumed according to Eq4. (Van Wijngaarden and al., 2011). This non-linear reaction rate reduces the liquid-phase concentrations of urea as a result of the bacterial activity.

$$r = \begin{cases} v_{\max} \cdot \frac{C_{\text{urea}}}{K_m + C_{\text{urea}}} \cdot \left(1 - \frac{t}{t_{\max}}\right) & \text{if } 0 \leq t \leq t_{\max} \\ 0 & \text{if } t > t_{\max} \end{cases} \quad \text{Eq4}$$

where v_{\max} represents the maximal reaction rate ($9.1 \cdot 10^{-5} \text{ kmol} \cdot \text{m}^{-3} \cdot \text{s}^{-1}$), t_{\max} the life span of bacteria ($6.12 \cdot 10^5 \text{ s}$), K_m the saturation constant ($0.010 \text{ kmol} \cdot \text{m}^{-3}$) and C_{urea} the concentration of urea.

Boundary conditions

A constant concentration is imposed to the feeding solution and no flow (i.e. no advection and diffusion) are imposed to the periphery of the sample and the axis of symmetry. An outflow boundary condition is imposed at the bottom of the sample. Chemical boundary conditions are summarized on Figure 3.



- 1: Constant concentration**
- 2: No-flow boundary condition**
- 3: Outflow boundary condition**

Figure 3. Geometry and chemical boundary conditions (no scale)

4. Results

The aim of the simulations was to verify the homogeneity of treatment prior to a series of laboratory tests. Thus, a parametrical study has been performed on the dispersion/diffusion coefficient to evaluate its impact on the homogeneity of treatment. An example of results is on Figure 4 presenting the evolution of the calcium content after 24h (a), 48h (b), 72h (c) and 96h (d) for a dispersion/diffusion coefficient of $4.3 \cdot 10^{-8} \text{ m}^2/\text{d}$.

The main observation is that the dispersion/diffusion ensures a homogenous treatment and prevents any preferential pathway under the drops of reactive water. Indeed, the isoconcentrations of calcite become almost horizontal after 96h and the calcium content is similar in the center of the sample or at the periphery.

A singularity is observed immediately at the entrance in the sample, as a result of the minimum of time necessary to perform the reaction. In practice, the samples are polished after treatment and this observation is not an issue.

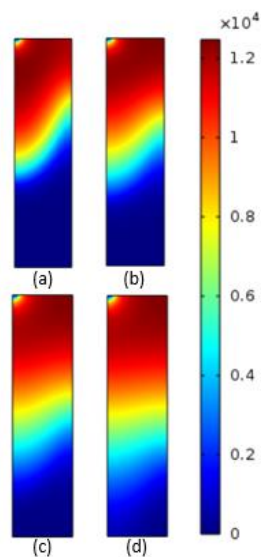


Figure 4. Evolution of calcium concentrations after 24h (a), 48h (b), 72h (c) and 96h (d)

These good theoretical results lead to the laboratory tests that confirmed the homogeneity of treatment, as observed on Figure 5 presenting sample after treatment and during an unconfined compression test.

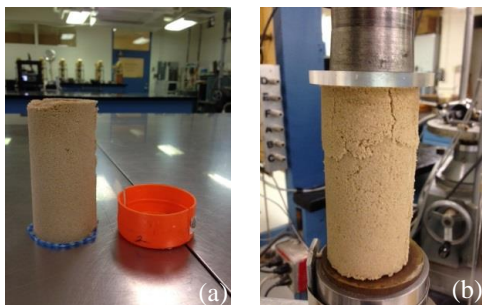


Figure 5. Example of calcified soil (a) before and (b) during unconfined compression test.

5. Conclusion

In this paper, a model of biocalcification in non-saturated was implemented in *Comsol Multiphysics* thanks to the *Subsurface flow module* and its application to Richard's equation and solute transport.

The simulations confirmed the homogeneity of treatment in non-saturated conditions as observed during our laboratory tests after preparation or unconfined compression tests. These simulations were mandatory to verify the conception of a new drip-fed irrigation system and prevented numerous adaptations.

6. References

1. Bachmeier, K.L., Williams, A.E., Warmington, J.R., Bang, S.S., 2002. Urease activity in microbiologically-induced calcite precipitation, *J. Biotechnol.*, **93**, 171–181.
2. Bang, S.S., Galinata, J.K., Ramakrishnan, V., 2001. Calcite precipitation induced by polyurethane-immobilized *Bacillus pasteurii*. *Enzym. Microb. Technol.*, **28**, 404–409.
3. Cheng, Cord-Ruwisch, Shahin, Cementation of sand soil by microbially induced calcite precipitation at various degrees of saturation, *Canadian Geotechnical Journal*, **50**, pp81-90 (2013)
4. DeJong, Mortensen, Martinez, Nelson, Bio-mediated soil improvement, *Ecological Engineering*, **36**, pp197-210 (2010)
5. Stocks-Fischer, S., Galinat, J.K., Bang, S.S., 1999. Microbiological precipitation of CaCO_3 , *Soil Biol. Bio-chem.*, **31**, 1563–1571 (1999)
6. Van Wijngaarden, W. K., Vermolen, F. J., Meurs, G. A. M., Vuik, C., 2011. Modelling Biogrout: A New Ground Improvement Method Based on Microbial-Induced Carbonate Precipitation, *Transport in Porous Media*, **87** (2), 397-420.
7. Whiffin, V.S., 2004. Microbial CaCO_3 Precipitation for the Production of Biocement, *Ph.D. Thesis, Murdoch University, Australia*.
8. Whiffin, V.S., van Paassen, L.A., Harkes, M.P., 2007. Microbial carbonate precipitation as a soil improvement technique. *Geomicrobiol. J.*, **25** (5), 417–423.