Modeling of Biocalcification in Non-saturated Conditions

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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 (CO(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.

\[
\text{CO(NH}_2\text{)}_2 + 2\text{H}_2\text{O} + \text{Ca}^{2+} \xrightarrow{\text{urease}} 2\text{NH}_4^+ + \text{CaCO}_3(s)
\]

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 pound 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.

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 pound 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.
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 hypotheses 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. $\text{Ca}^{2+}$ 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 $\text{Ca}^{2+}$ 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.

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

where $c$ represents the dissolved concentration (mol/m$^3$), $\theta$ the volume fluid fraction (-), $D_L$ the hydrodynamic dispersion tensor (m$^2$/d) and $R_L$ the reaction rates (mol/(m$^3$.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.

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

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

![Figure 3. Geometry and chemical boundary conditions (no scale)](image)

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

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 $4.3 \times 10^{-8}$ m$^2$/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 during our laboratory tests after preparation or unconfined compression tests.

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