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Research paper

Effects of clay's chemical interactions on biocementation

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ABSTRACT

Biocementation, or microbially induced calcite precipitation (MICP), is a technique currently appraised for the improvement of sandy soils. These types of soils are good candidates for MICP-based soil improvement due to their relatively large voids size associated to high permeability and room available for bacteria colonization. Only few studies use soils with controlled percentages of clay fraction (up to 20%), in which MICP is still efficient. However, chemical activity in the clay fraction is not considered, but may be relevant for biocementation because the clay minerals react with the feeding solution (due to pH and the presence of calcium ions). In this paper, the effect of clay chemical interactions on biocementation is investigated considering samples of a uniformly graded sand and samples of the same sand to which kaolin clay was added to reduce the porosity to half. The changes on some physical properties of these artificial soils achieved after treatment were evaluated in 1) saturated samples through changes in compressibility and permeability, and 2) in partially dried samples through changes in tensile strength. The results of the tests are discussed considering the presence of a clay and calcium carbonate coating, both bonding the grains of sand. The presence of clay minerals has two main effects: (i) an increase on the compressibility due to osmotic effects that reduce clay stiffness and (ii) a reduction in the permeability associated to pore size reduction. MICP treatment per se appears not to affect significantly this behavior. Although the tensile strength increases for the sand samples with clay, the increment is larger after MICP, which could be explained by apparent cohesion and physical connections from calcium carbonate minerals. The results allow concluding that chemical interactions between the feeding solution and the clay minerals must be considered when using MICP in soils containing them and, for this reason, this treatment in clayey soils is more complex than in sands without significant percentage of clay.

1. Introduction

Biocementation, or microbially induced calcite precipitation (MICP), is a soil treatment technique based on bacterial activity that alters the hydraulic and mechanical properties of porous media (see for example DeJong et al., 2013). MICP provides adequate conditions to specific bacteria, living in the soil, hydrolyses urea ($(NH_2)_2CO$) (Eq. (1)). The ammonium (NH_4^+) released from urea hydrolysis results in pH rise that promotes the precipitation of calcium carbonate ($CaCO_3$) (Eq. (2)).

$$(NH_2)_2CO + 2H_2O \rightarrow 2NH_4^+ + CO_3^{2-} \quad (1)$$

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

Calcium carbonate formed, also known as biocement, accumulates in the soil pores mainly between the grains (DeJong et al., 2006), therefore clogging the pores and reducing permeability. In addition, the biocement introduces stiffness and strength because it is a physical connection (named as bond in this paper). Calcium carbonate can precipitate in three main polymorphs: vaterite, aragonite and calcite (Wei et al., 2015). Environmental conditions significantly influence the type of polymorph formed (Teng et al., 1998; Davis et al., 2000), but the type of microorganisms must also be considered (Cañveras et al., 2001; Dupraz and Visscher, 2005; Wei et al., 2015). Wei et al. (2015) found that vaterite is the main form of calcite produced by the ureolytic gene of the bacterial species used in this work, *Sporosarcina pasteurii*, a result also found by Cardoso et al. (2017) in XRD tests.

Previous studies mainly focused on improving sands and silty sands. Concerning hydraulic properties, permeability can be reduced about one order of magnitude (Whiffin et al., 2007; Cheng et al., 2013; Zamani and Montoya, 2016). Kirkland et al. (2017) found a final porosity 85% of the initial porosity of a silty sand. Concerning the mechanical properties, strength increment is evaluated mainly through unconfined compression tests (Al Qabany and Soga, 2013, among

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others), however there are several references presenting other tests, such as direct shear and triaxial drained tests (Montoya and DeJong, 2015; Feng and Montoya, 2014, 2015) and Brazilian splitting tests (Van Paassen, 2009). As far the authors knowledge, data on one-dimensional oedometric compression tests are scarce, however there are some studies (Lee et al., 2013; Lin et al., 2015; Cardoso et al., 2016) focused on compressibility changes due to biocementation.

The improvement on the hydraulic and mechanical properties depends directly on the amount of biocement present to form bonds. Bond distribution is not homogeneous and represents the main source of inconsistencies in experimental data (Al Qabany and Soga, 2013). Calcium carbonate percentages (in weight of calcium carbonate by weight of dry soil) found in the literature range from ~1% (DeJong et al., 2006) to 25% (Van Paassen, 2009). These values depend on several parameters as the grading size distribution of the soil, injection conditions of both bacteria and feeding solution, and if are determined by chemical analysis, or through indirect methods (for example, through changes on porosity or shear wave velocity).

Several existing laboratorial studies aimed to understand the most adequate ways to perform this treatment in soils, mainly by determining bacteria concentrations and establishing feeding composition and frequency (DeJong et al., 2006; Al Qabany et al., 2012; Al Qabany and Soga, 2013; Montoya et al., 2013; Terzis et al., 2016; Dhami et al., 2016, among others). The vital space for bacteria to survive has been investigated by considering different grading size distributions and void ratio, because: (i) they affect voids volume and soil saturated permeability, and therefore nutrient access to the bacteria living in soil pores (Cheng et al., 2013; Cheng and Cord-Ruwisch, 2014; Terzis et al., 2016; Cardoso et al., 2016), or (ii) simply are associated to lack of space for bacterial growth (Stocks-Fischer et al., 1999; Mitchell and Santamarina, 2005; Phadnis and Santamarina, 2011). Sandy soils are investigated because these physical properties are naturally achieved, because soil voids have size proportional to grain size. Studies using sandy soils prepared with controlled clay percentages (Cheng and Shahin, 2015) or on soils with some clay percentage only focus the analysis of hydraulic and mechanical properties.

The effect of high salt concentration of the feeding solution on MICP efficiency is also investigated, because this may result in inhibitory effects on microbial activities (Whiffin et al., 2007; De Muynck et al., 2010; Dhami et al., 2016). However, the chemical interaction between the feeding solution and bacteria and the soil (mainly sand) is not considered. These chemical interactions are investigated in the current paper because they are relevant in clayey soils, as clay minerals react with the percolating fluid. This interaction has consequences on the behaviour of clayey soils which, as well known, are affected by the chemical nature of the percolating fluid and can exhibit deformations (osmotic consolidation) and significant changes in their mechanical response (Di Maio et al., 2004; Amarasinghe et al., 2011). Even if this chemical interaction is well known in clays, it has not been study in the context of biocementation. In this case, changes in clay soil behavior due to chemical interaction with the percolating fluid can be explained by ion adsorption by clay minerals and consequent changes in the distance between parallel clay platelets (double layer theory, Mitchell and Soga, 2005) due to repulsive or attractive forces.

In this paper, the effect on biocementation of the presence of clay soil partially filling the pores between the particles of sand is investigated, aiming to understand the relevance of the clayey minerals in this process. Samples of an uniformly graded sand (named as sand) and samples with the same amount of sand but to which a given amount of clay soil was added (named as sand-kaolin) were prepared (illustration in Fig. 1). The weight of sand was the same for both kind of samples, fixing the porosity from the arrangement of sand grains. For the sand-kaolin samples, a clay soil was added to reduce the porosity to half. Both samples were treated with bacteria and feeding solution (pH = 9), but for checking the chemical effects (due to pH and the presence of calcium) the tests on the samples with clay soil were repeated for samples to which only feeding solution was added (pH = 9). Another set with samples prepared only with distilled water (pH = 7) was used as reference. They allowed investigating the effect of the clay presence when comparing the sand with the sand-kaolin samples.

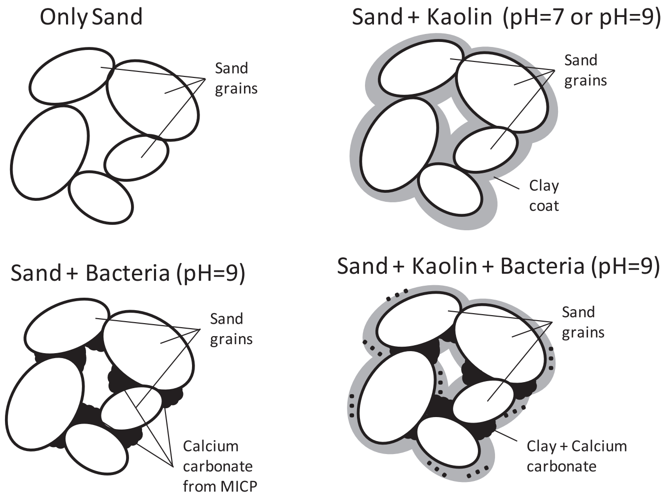

Fig. 1. Physical interpretation of the samples prepared using sand and sand-kaolin, considering the coating introduced by the clay soil and calcium carbonate bonds.

The mechanical properties of the different materials were evaluated in oedometric compression and in Brazilian splitting tests. Complementary information was obtained from mercury intrusion porosimetry tests (MIP) and scanning electron microscope (SEM) images. The oedometer tests provided information about oedometric compressibility and were used to estimate the evolution of the saturated permeability. The differences in both properties can be explained by changes in pore size distribution and osmotic effects such as pH and calcium interaction with the clay minerals, as well as by the presence of calcium carbonate filling the soil pores. This physical effect is illustrated in Fig. 1. The Brazilian tests provided information about the presence of a clay and calcium carbonate coat bonding the grains, which correspond to physical connections (bonds) that may break under tension. The differences in testing all cases provided indirect information about the relevance of pore clogging and the nature of the bonds, as well as if MICP may be affected by the clay presence.

2. Materials and experimental setup

2.1. Bacteria

The bacteria *Sporosarcina pasteurii* (American Type Culture Collection, strain #11859) was grown in liquid medium composed by 20 g/L yeast extract, 10 g/L of ammonium sulphate and 0.13 M Tris buffer pH = 9.0, at 37 °C under aerobic condition. The concentration adopted for the treatments was ~10^9 cells/mL, which corresponds to an optical density at 600 nm (OD600) of 1. The feeding solution was prepared with 0.5 M of urea, 0.5 M of calcium chloride (calcium source) and 1:10 diluted culture medium, with Tris pH = 9.

2.2. Soils used

The samples tested were prepared using sand and white kaolin clay. The pH values of the two materials when mixed with distilled water were around 7.0.

A commercial river sand (reference APAS20) was used with an almost uniform grading size distribution (diameters between 0.4 and 2 mm) (Fig. 2). The solid volumetric weight of this sand, mainly quartz mineral, is 26.4 kN/m^3. The maximum and minimum voids ratio achieved are 0.938 and 0.573, determined for the dry material using