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Manuscript title: A numerical inspection on the squeezing test in active clays

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Abstract

Squeezing tests are performed to obtain liquid samples from soil to characterise the chemical composition of the aqueous saline solution in its pores. However, the representativeness of the extract obtained from active clays has been debated, as a mixture of macrostructural and microstructural solutions is likely to be obtained. To analyse this question, given the complexity and the strong hydro-chemo-mechanical coupling of the processes that occur during a squeezing test, it is of interest to simulate the test using a conceptual model able to characterise the processes involved in active clays. This type of simulation is performed in this study, showing that the chemo-mechanical interaction between macrostructure and microstructure is unavoidable. However, in saturated soils with low dry densities, most of the extracted water is directly obtained from the macrostructure, and the extract can be considered representative of the macrostructural solution. With increasing dry density, the mixing increases, and the representativeness of the extract may be compromised. In unsaturated soil with a low water content and high dry density, an important mass exchange occurs between the macrostructure and microstructure. If the test is viable (i.e., sufficient solution volume can be obtained), the representativeness of the extract must be carefully evaluated.

Keywords: Bentonite; expansive soils; squeezing test; chemo-mechanical coupling; model tests; numerical modelling
INTRODUCTION

Characterising the chemical composition of the aqueous solution in the soil pore space is a matter of interest for both geochemical and geomechanical studies, particularly for active clays, given the strong hydro-chemo-mechanical coupling associated with these materials (Di Maio, 1996; Studds et al., 1998). Thus, obtaining samples that are representative of the chemical composition of the interstitial solution is essential. Given the low hydraulic conductivities of active clays and their ability to develop high suction, such samples are not easily obtained. One of the procedures used to obtain samples is the squeezing technique (for example, Muurinen et al., 2004; Arifin & Schanz, 2009). However, doubts have been raised concerning the “true” representativeness of the solution obtained. In addition to the changes in the chemical composition of the liquid extract that can be caused by temperature changes and contact with atmospheric gases, the very high pressure applied (50 MPa, Arifin & Schanz, 2009, or even higher, Mazurek et al., 2015) can mobilise liquid from both large pores and interlayer spaces (Fernández et al., 2014; Muurinen et al., 2007), which may mix and thus distort the representativeness of the extract. Analysing this potential mixing is the main objective of this paper.

Arifin & Schanz (2009) experimentally investigated mixing and analysed the effect of the soil specific gravity and the initial values of the void ratio and degree of saturation on the squeezing pressure, defined as the pressure that must be applied to the soil to obtain a “first extract drop”, which is assumed to be characteristic of the soil-pore solution in high plasticity clays (Arifin & Schanz, 2009). By installing electrodes inside MX-80 bentonite specimens, Muurinen & Carlsson (2007) verified that the values of pH (acidity) and Eh (redox potential) measured by the electrodes in a specimen with an initial low dry density were equal to those values of the squeezed pore solution when the compressed specimen reached a density of 1.5
In addition, Fernández et al. (2014) and Mazurek et al. (2015) investigated the more compact Opalinus Clay from the Mont Terri underground rock laboratory in Switzerland. Both studies identified a threshold squeezing pressure (175 MPa according to Fernández et al., 2014, and 200 MPa for Mazurek et al., 2015) below which an extract is considered to be closely representative of the in situ composition of the free pore water.

These contributions provide empirical evidence of a threshold above which mixing is relevant. However, a conceptual model that facilitates a better understanding of the mechanisms and processes that are developed in a squeezing test is of interest. Muurinen et al. (2004, 2007) proposed a conceptual model to reproduce the squeezing of an MX-80 bentonite. They confirmed that single porosity models do not reproduce consistently the performed tests, indicating the need for a double porosity approach. Therefore, the double porosity approach used in the hydro-chemo-mechanical model of Navarro et al. (2017c) provides a basis for the numerical inspection performed in the present study. The model assumes a simplified geochemical configuration, considering only Ca$^{2+}$, Na$^{+}$ and Cl$^{-}$ ions together with the clay mineral. However, the verification and validation exercises conducted up to date (Navarro et al., 2016, 2017a, 2017b, 2017c) (see the “Conceptual and Numerical Model” section) have proven the ability of the model to reproduce the stress-strain behaviour of an MX-80 bentonite under different chemo-mechanical loads. Thus, it has been used to simulate ten squeezing tests with different salinity, saturation and dry density conditions, as well as with different squeezing pressures and loading rates. The results obtained highlight the complexity and coupling of the processes developed during the squeezing tests, and revealed that a certain mixture occurs in all cases (as indicated by Muurinen et al. 2004). In addition, the simulations show that, when squeezing extracts from unsaturated compacted
specimens, the mixing process is so intense and complex that the representativeness of the extract is compromised.

Prior to presenting the results and after describing the materials and conditions assumed in the tests, the main characteristics of the model are outlined, which is described in more detail in Navarro et al. (2017c).

**MATERIALS AND METHODS**

The material considered for the numerical simulations was the natural sodic MX-80 bentonite modelled by Navarro et al. (2017a, 2017b, 2017c). This material has similar properties to the material Be-Wy-BT007-1-Sa-R described by Kiviranta & Kumpulainen (2011). The main mineralogical, chemical and physical properties of this bentonite are listed in Table 1.

The geometry and boundary conditions adopted are summarised in Fig. 1. The modelled specimens were cylindrical with a diameter and a height of 20 mm, similar to those employed by Muurinen et al. (2004) and Arifin & Schanz (2009). In oedometric conditions (both lateral strains, flow and transport were assumed to be null), a vertical load was applied on top of the sample in the form of a ramp function, which started in a zero load and reached a maximum value of the squeezing pressure $\Delta \sigma$ after a time $t_{\Delta \sigma}$ (Table 2). The load remained stationary at its maximum value from this point until the end of the simulation. Lateral friction effects were not considered in the simulations. The top boundary of the specimen is impervious to water flow and ion transport. The bottom was also assumed impervious for macrostructural degrees of saturation less than one. However, when this boundary is fully saturated, the macrostructural liquid pressure is assumed to be equal to the atmospheric pressure (seepage or free surface boundary condition, Franke et al., 1987; Scudeler et al., 2017). In addition, a third-type or flux-type boundary condition (van Genuchten and Parker, 1984) is applied to the
macrostructural transport of chemical species in saturated conditions. Therefore, the flow and transport boundary conditions at the sample bottom are defined as:

\[
\begin{align*}
\text{if } S_{RM} < 1: & \begin{cases}
q_M = 0 \\
I_i = 0
\end{cases} \\
\text{otherwise:} & \begin{cases}
P_L = P_{atm} \\
I_i = C_{i,M} q_M
\end{cases}
\end{align*}
\] (1)

where \( S_{RM} \) is the macrostructural degree of saturation, \( q_M \) is the macrostructural water seepage, \( P_L \) is the liquid pressure, \( P_{atm} \) is the atmospheric pressure, and \( I_i \) and \( C_{i,M} \) are, respectively, the mass flow with regard to the soil skeleton displacement and the molar concentration in the macrostructural water of the i-th ion (Cl\(^{-}\), Na\(^{+}\) and Ca\(^{2+}\)).

As indicated in the Introduction, ten squeezing tests were simulated. Like in the tests performed by Muurinen & Carlsson (2007) and Muurinen et al. (2004), saturated conditions were adopted in nine simulations (see Table 2). In eight of those simulations, an open soil with an initial dry density \( \rho_D \) of 0.7 g/cm\(^3\) (minimum value considered by Muurinen et al., 2004) was modelled. In the remaining saturated simulations, the value of \( \rho_D \) was twice as large, 1.4 g/cm\(^3\). This way, the effect of soil density on its response was assessed. In addition, to analyse the effect of soil water content, the squeezing of an unsaturated as-compacted specimen, with a gravimetric water content of 17\% and the same dry density of 1.4 g/cm\(^3\) (T4, Table 2), was simulated. The sensitivity of the results to the density of microstructural water \( \rho_m \) was assessed by using a function for it based on Jacinto et al. (2012) (see Appendix B) in one of the eight saturated simulations of an open soil (T2, Table 2), while using \( \rho_m = 1 \) g/cm\(^3\) in the rest of simulations. The sensitivity to the squeezing pressure (simulations T1b1 and T1b2) and to the loading rate (simulations T1a1 and T1a2) was also studied. Finally, to illustrate the effect of the initial salinity of the soil, soils with an initial macrostructural salts concentration of 1 g/L (T1c1, Table 2) and of 35 g/L (T1c2, Table 2) were simulated, in
contrast with the 10 g/L assumed in the rest of cases. The 1 g/L concentration represents a low salinity water. The 10 g/L concentration represents a brackish-saline water simulant for Olkiluoto, the site of the Onkalo deep geological repository for spent nuclear fuel in Finland, while the 35 g/L concentration represents the maximum expected salinity of the groundwater at the repository depth (Hellä et al., 2014).

**CONCEPTUAL AND NUMERICAL MODEL**

As noted in the Introduction, the conceptual framework used behind the simulations is based on the double porosity chemo-mechanical model proposed by Navarro et al. (2017c). Since the present article is based on its application, this section presents a synthesis of its main features (complemented by Appendices A to D) and some examples of application that illustrate the capacity of the model to simulate squeezing tests. Further details on the model and application can be found in Navarro et al. (2017a, 2017b, 2017c).

As it is common in the analysis of the behaviour of compacted bentonites (Alonso et al., 2005; Gens, 2010; Guimarães et al., 2013; Lemaire et al., 2004; Nowamooz et al., 2009; Sedighi & Thomas, 2014; Vilarrasa et al., 2016; Xie et al., 2004), the model defines macrostructure and microstructure as the inter-aggregate void space and the intra-aggregate void space, respectively. The total void volume per mineral volume $e$ is defined as (see, for example, Sánchez et al., 2005, and Musso et al., 2013):

$$
e = e_m + e_M$$  \hspace{1cm} (2)

where $e_m$ is the microstructural void ratio (volume of intra-aggregate voids per volume of mineral), and $e_M$ is the macrostructural void ratio (volume of inter-aggregate voids per volume of mineral). If $e_M$ is greater than $e_{M_{\text{min}}}$ (which is assumed to be equal to 0.01) the definition of the variation in $e_M$, and the characterisation of the macrostructural stress-strain behaviour is based on the "Barcelona Basic Model" (Alonso et al., 1990). When $e_M$ is less
than \( e_{\text{Mmin}} \), then the exponential approximation of the bulk modulus increment defined in Fig. A.1 was adopted. The definitions and values of the parameters used in the mechanical model of the macrostructure are included in Appendix A, which also describes how to obtain the microstructural strains from the variation of \( e_m \).

To determine \( e_m \), the state surface represented in Fig. 2 is used, where \( s_{mS} \) defines the “structural” suction of the microstructure. This suction term is equivalent to the thermodynamic swelling pressure introduced by Navarro et al. (2017a). For the MX-80 bentonite analysed in this study, \( e_{mR} \) (the remaining microstructural void ratio in dry conditions) is 0.093 (Navarro et al., 2015). Considering the equivalency between suction and water potential (Yong, 1999), \( s_{mS} \) defines the potential that is related to the organisation of the microstructure, which is characterised by \( e_m \). Although several authors noted that macrostructural and microstructural water may not be in equilibrium (for instance, Ferrage et al., 2007; Montes-H et al., 2003; Musso et al., 2013), Navarro et al. (2015) confirmed that the flow in the bentonite analysed in the present work is slow enough to assume a fast macrostructure-microstructure mass exchange and thus equilibrium. As detailed in Appendix B, in that case \( s_{mS} \) can be calculated as:

\[
s_{mS} = p + s_M + s_{MO} - \Delta s_{mNCCI}
\]

where \( p \) is the net mean stress, defined as the mean stress minus the gas pressure \( P_G \), and \( s_M \) is the macrostructural matric suction, identified as the capillary suction, and therefore calculated as the difference between \( P_G \) and the liquid pressure \( P_L \); \( s_M = P_G - P_L \). The function \( \Delta s_{mNCCI} \) is the increase in microstructural suction due to the contribution to the chemical potential of ions in excess of the cation exchange capacity \( CEC \) (the non-charge compensating ions or “extra” microstructural salinity), and \( s_{MO} \) is the osmotic suction of the macrostructural water. The terms \( s_{MO} \) and \( \Delta s_{mNCCI} \) are functions of the concentrations of
calcium, sodium and chloride in the macrostructure and in the microstructure, respectively (Appendix B).

Given the fast kinetics of the ion exchange in motmorillonite (Sparks, 2003), the Donnan equilibrium approach (Helfferich, 1962) is adopted, and therefore, the macrostructural and microstructural concentrations are related by the partition function $B$. In addition to these three equations, an additional electroneutrality equation at each of the two structural levels is considered. Then, the geochemical system has seven unknowns (six concentrations and $B$) and five equations; that is, there are two degrees of freedom. To solve them, the molar concentration of calcium in the macrostructure $C_{\text{Ca,M}}$ and that of chloride in the microstructure $C_{\text{Cl,m}}$ are adopted as the main unknowns (Appendix B).

Since isothermal conditions (at 25°C) and a constant gas pressure that is equal to the atmospheric pressure are assumed, these two concentrations and $P_L$ constitute the state variables of the flow and transport model described in Appendix C. These calculations are coupled with the characterisation of the soil skeleton displacement field $\mathbf{u}$ and the stress tensor to introduce the value $p$ into Eq. (3). Thus, the flow and transport mass balance equations and the mechanical equilibrium equation must be solved in a coupled manner. To this end, the formulation described in Appendices A to C was implemented (see Appendix D) using the “multiphysic” feature in COMSOL Multiphysics (Navarro et al., 2017c), which is a multiphysics partial differential equation solver based on the finite element method with Lagrange multipliers (COMSOL, 2015). The developed numerical tool was employed to simulate the squeezing tests defined in the previous section and to obtain the results presented in the following one.

Despite the simplified abstraction of the geochemistry of the system (including only $\text{Ca}^{2+}$, $\text{Na}^+$ and $\text{Cl}^-$ and clay), the model produced good results. This is illustrated in Fig. 3(a),
adapted from Navarro et al. (2017c), which plots the results of the simulation of a vertical free swelling process of an MX-80 bentonite. Despite the large strains occurred (obtaining water contents over 1000%), the model is able to reproduce the experimental results. The fit of the model results to the experimental ones is also noteworthy in the swelling pressure tests in Fig. 3(b) (tests by Karnland et al., 2005, 2006). In them, the salts concentration varies significantly. Fig. 3(b) plots the correlation between the model and the experimental final swelling pressure values. However, the model has also been confirmed to reproduce the time evolution of the swelling pressure. These cases are very demanding on the model, especially regarding its ability to model the water mass exchange between macrostructure and microstructure. The quality of the results obtained provides confidence in its application to study the potential mixing between macrostructural and microstructural water in squeezing tests.

RESULTS AND DISCUSSION

Figs. 4 and 5 show the evolution of the mean molar concentrations of the ions and the ionic strength, IS, in the extract (which is assumed to be a perfectly mixed system). In addition, the initial value of these variables in the macrostructure, which are the values aimed to be characterised with the squeezing, have been represented in a dashed line for comparison. As can be seen in Fig. 4(a), the effect of the loading rate is not significant. The hydraulic conductivity of bentonite is so small that nearly all the samples behave as undrained even for the higher loading rate, $\Delta \sigma = 10$ min. This fact is illustrated in Fig. 6, which shows the differences between the point A (sample top, see Fig. 1) and points B and C, where the liquid pressure is equal to the applied load.

On the other hand, the effect of the value of the squeezing pressure is relevant (Fig. 4(b)). Greater pressures lead to greater reductions of $e_m$ (Fig. 7), more presence of microstructural
fluid in the extract (Fig. 8) and thus more distortion introduced in the test: the composition of the extract differs more from the initial composition of the macrostructural water. The role of the initial salts concentration is also relevant. The greater it is, the more representative the extract is of the real (initial) macrostructural salinity (Fig. 5).

Assuming the density of microstructural water as 1 g/cm$^3$ does not seem to significantly distort the results. Fig. 9 illustrates that using the function based on Jacinto et al. (2012) (see Fig. B.1, Appendix B) for $\rho_m$ produces results similar to those using $\rho_m=1$ g/cm$^3$ (the differences are below 3.4%).

The extract salinity (characterised by its ionic strength) decreases with time with respect to the initial macrostructural value, as derived from Figs. 4 and 5. However, the difference in ionic strength is lower than 9.8% (Test T1) for the lowest dry density simulated, with a proportion of the extract from the microstructure lower than 16% (Fig. 8). For this dry density, the extract seems to remain largely representative. However, the capacity of numerical simulation to explain processes illustrates that, even if the extract is not very distorted, the behaviour of the system is not simple.

Although the extract salinity decreases with time, its sodium concentration increases slightly, because the decrease in its chloride concentration is lower than twice the decrease in its calcium concentration (Fig. 4). The complexity of the soil response becomes fully apparent in an aspect not usually considered when analysing squeezing tests: the high hydro-mechanical heterogeneity induced during the test. As noted when discussing the liquid pressure values earlier in this section, the small hydraulic conductivity causes different effects at different locations. Near the draining edge (point A of Fig. 1), the water overpressure rapidly dissipates, and the entire applied load results in increased effective stress (Fig. 6). In the inner part of the specimen (points B and C, Fig. 1), this dissipation is minimal, and the load results
in increased pore water pressure, maintaining a constant effective stress at the considered times. Therefore, the macrostructural void ratio is significantly reduced at the edges while exhibiting minimal variation in much of the specimen, as shown in Figs. 7 and 10. Both figures indicate a similar trend in the microstructural void ratio. In saturated conditions, Eq. (3) can be written as:

\[ s_{mS} = p' + s_{MO} - \Delta s_{mNCCI} \]  

where \( p' \) is the effective mean stress. Given that the hydro-mechanical load (\( p' \)) becomes significantly higher than the chemical load (\( s_{MO} - \Delta s_{mNCCI} \)) (Fig. 6), both \( s_{mS} \) and \( e_m \) are fundamentally controlled by \( p' \), which explains the parallels between the evolution of \( e_M \) and the evolution of \( e_m \) (however, given the large value of \( e_M \) in the low dry density bentonite analysed, the variation in the macroporosity is considerably higher than the variation in the microporosity). Consequently, the variability in the chemical composition of the extract can be considered to be fundamentally induced by the temporal evolution of the macrostructural consolidation process. Nevertheless, this assumption is not entirely true.

Fig. 11 shows the variation in the ion concentration along the length of the specimen after ten minutes of testing. The concentrations of calcium and sodium ions in the microstructure show large variations. A reduction in \( e_m \) increases the surface charge density, Eq. (B.10), which increases the concentrations of \( \text{Ca}^{2+} \) and \( \text{Na}^+ \) and decreases the concentration of \( \text{Cl}^- \) to maintain the electroneutrality in the microstructure, Eq. (B.9). The decrease in the concentration of \( \text{Cl}^- \) is barely noticeable in Fig. 11 because the \( \text{Cl}^- \) concentration in the microstructure (initially 31 mM) is significantly smaller than the concentrations of \( \text{Na}^+ \) (initially 623 mM) and \( \text{Ca}^{2+} \) (initially 995 mM). However, as shown in Fig. 12, the decrease in \( \text{Cl}^- \) is significant in relative terms. During squeezing, water can be extracted from the microstructure (Fig. 8), and the deformation of the microstructure changes the surface charge.
density and the chemical composition of the macrostructural and microstructural solutions, and therefore changes the extract. The medium is not inert, and the extraction of water and solutes is not proportional. The chemical composition of the extract is the system response to a boundary problem in which numerous coupled processes are developed. The composition of the extract can significantly deviate from the initial composition in the macropores and does not even have to be associated with any characteristic value of the medium.

Nevertheless, in a specimen with a structure as open as that of tests T1 to T2 ($\rho_D = 0.7$ g/cm$^3$), the microstructure does not have a strong influence. As noted above (see Figs. 4, 5 and 9), the chemical composition of the extract is similar to the initial chemical composition in the macrostructure. This result was not obtained in the T3 test, which also considered a saturated specimen but with an initial dry density of 1.4 g/cm$^3$. In this case (Fig. 13), as squeezing progressed, the chemical composition of the extract significantly differed from the initial chemical composition in the macrostructure. As the initial macrostructural void ratio (= 0.251 in T3) was considerably smaller than the initial macrostructural void ratio in tests T1 to T2 (= 2.433), 60 seconds after the start of the test, $e_M$ became approximately nil at the top of the specimen (Fig. 14). After this situation, almost all of the extract was obtained from the microstructure, as reflected in the trend change from approximately 60-70 seconds in Fig. 13, and the difference between the obtained extract and the initial chemical composition of the macrostructural solution increases.

The time at which this trend change occurs, which can be understood as the threshold where the mixing between the solution extracted from the macrostructure and that extracted from the microstructure becomes relevant, is not easily determined. This value depends not only on the initial dry density of the specimen and the macrostructural deformability of the soil but also on the microstructural volumetric behaviour, as the shrinkage of the microporosity may
enlarge the macroporosity, and on the flow and transport properties of the material; that is, on the constitutive hydro-chemo-mechanical properties of the soil. Therefore, it is difficult to obtain general guidelines for determining either the magnitude of the load or the speed with which it should be applied to decrease the effect of $e_M$ becoming practically zero.

The process is even more complex if started from a partially saturated compacted specimen, such as in test T4. In this case, when the macrostructural void ratio was reduced, Fig. 15, the macrostructural degree of saturation increased, decreasing $s_M$ and increasing $P_L$, as shown in Fig. 16. Consequently, $s_{mS}$ decreased, Eq. (3), and $e_m$ increased, as shown in Fig. 15. Thus, in the initial test phase, the water in the macrostructure was transferred to the microstructure, increasing the microstructural void ratio. When the macrostructure was depleted, $e_m$ decreased but only slightly. By increasing $p$ and decreasing $s_M$, $s_{mS}$ exhibits minimal variation, as stated in Eq. (3) and shown in Fig. 16. In this case, the viability of squeezing must be assessed because the volume of liquid extracted is very small and is obtained from the microstructure.

**CONCLUSIONS**

Given the complexity of the hydro-chemo-mechanical coupled processes that occur in the squeezing of an active clay, simulating this test using a numerical model is useful for reproducing, identifying and understanding the processes involved. This study uses the conceptual model from Navarro et al. (2017c), which has a demonstrated ability to simulate the main behaviour characteristics of these materials. The application of this model showed that the geochemical coupling between the macrostructure and microstructure of the active clay is significant, even in saturated soils with low dry densities, in which a substantial amount of the extracted water is obtained from the macrostructure. This coupling is more significant at a higher dry density, even while maintaining saturated conditions. In this case,
when the macrostructural void ratio becomes approximately zero at the draining edges as a consequence of the squeezing-induced consolidation, the extracted liquid solution is predominantly from the microstructure. However, before this blockage, the extract can be considered representative of the initial composition of the macrostructural solution, similar to the less dense specimens. This conclusion is not true for partially saturated soils compacted to a high density. In this case, the water in the macrostructure is transferred to the microstructure before the extract is obtained. Given the processes that develop before obtaining the extract, it is advisable to evaluate the representativeness of the extract, provided that it is large enough to be analysed.

The extract can be expected to be stable and chemically representative only in sufficiently open soils. In active clays under other conditions, generalisations are difficult; thus, the results of squeezing tests must be carefully interpreted.

ACKNOWLEDGEMENTS

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APPENDIX A. MECHANICAL MODEL

In the monotonic compression paths that are analysed in this paper, a simplified additive structure was assumed to define the total soil strain increment vector $d\varepsilon_{\text{TOT}}$ (Navarro et al., 2016):

$$
d\varepsilon_{\text{TOT}} = d\varepsilon_{\text{M}} + d\varepsilon_{\text{m}}. $$

(A.1)

The vector $d\varepsilon_{\text{M}}$ defines the strain increment caused by macrostructural deformation. As noted in the Conceptual and Numerical Model Section, if $e_{\text{M}}$ is greater than $e_{\text{Mmin}}$ (taken as 0.01), then the “Barcelona Basic Model” (BBM; Alonso et al., 1990) was used. Therefore, the mechanical behaviour of the soil macrostructure was assumed to be described by two “significant stresses”: the macrostructural matric suction $s_{\text{M}}$ and the net stress $\sigma''$. The tensor field $\sigma''$ is defined as $\sigma'' = \sigma - P_{\text{G}} \cdot \mathbf{m}$, where $\mathbf{m}$ is the vector form of the Kronecker delta (the engineering or Voigt notation is used for both stress and strain tensors), and $\sigma$ is the total stress that defines the mechanical equilibrium equation:

$$
\nabla \cdot \sigma - \rho g \nabla z = 0
$$

(A.2)

where “$\nabla \cdot$” is the divergence operator, $\rho$ is the average soil density, $g$ is the gravitational acceleration, “$\nabla$” is the gradient differential operator and $z$ is the vertical coordinate (upwards oriented). From $\sigma''$, the net mean stress $p (p = \mathbf{m}^\top \cdot \sigma''$, where “$\top$” indicates the transpose operator) is computed. The net mean stress and $s_{\text{M}}$ define the increment of the elastic volumetric strain $d\varepsilon_{\text{MV}}^e$ in the macrostructure:

$$
d\varepsilon_{\text{MV}}^e = \frac{dp}{K_p} + \frac{ds_{\text{M}}}{K_s}
$$

(A.3)

where the stress, $K_p$, and suction, $K_s$, bulk moduli are computed as:

$$
K_p = \frac{p (1 + e)}{\kappa_p}; \quad K_s = \frac{(\kappa_{\text{ATM}} + P_{\text{ATM}}) (1 + e)}{\kappa_s}
$$

(A.4)
where $\kappa_p$ and $\kappa_S$ are, respectively, the elastic stiffness for changes in net stress and suction.

Their values, included in Table A.1, were identified by Navarro et al. (2016).

This paper considers yield only by contact with the loading-collapse $F$ surface to compute the increment of macrostructural plastic strains $d\varepsilon_M^p$, where $F$ is:

$$F = q^2 - M^2 (p + p_s)(p_o - p) = 0$$

(A.5)

where $q$ is the von Mises stress, $M$ is the slope of the critical state line (a material parameter; Table A.1), $p_o$ is the net mean yield stress at the current suction, and $p_s$ is the increase in tensile strength with suction, calculated as:

$$p_s = k s_M$$

(A.6)

The material parameter $k$ is included in Table A.1. The value of $p_o$ is given by the expression:

$$p_o = p_C \left( \frac{p_o^*}{p_C^*} \right)^{\frac{\lambda(0) - \kappa_p}{\lambda(s_M) - \kappa_p}}$$

(A.7)

where the reference stress $p_C$ is defined in Table A.1, and the evolution of the saturated pre-consolidation stress $p_o^*$ is calculated from the macrostructural plastic strain increment, $d\varepsilon_{MV}^p$, using the hardening law:

$$dp_o^* = \frac{(1 + \varepsilon)}{\lambda(0) - \kappa_p} d\varepsilon_{MV}^p$$

(A.8)

The function $\lambda$ defines the slope of the virgin compression curve at macrostructural suction $s_M$. It is computed as:

$$\lambda(s_M) = \lambda(0) \left[ r + (1 - r) \exp(-\beta s_M) \right]$$

(A.9)

where $\lambda(0)$, $r$ and $\beta$ are material parameters (Table A.1).
Table A.1. Macrostructural stress-strain parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k$</td>
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</tr>
<tr>
<td>$\kappa_p$</td>
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</tr>
<tr>
<td>$\kappa_S$</td>
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<tr>
<td>$p_{\text{REF}}$ (kPa)</td>
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</tr>
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<tr>
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<td>$\lambda(0)$</td>
<td>0.15</td>
</tr>
<tr>
<td>$r$</td>
<td>0.8</td>
</tr>
<tr>
<td>$\beta$ (kPa$^{-1}$)</td>
<td>$2.0 \cdot 10^{-5}$</td>
</tr>
<tr>
<td>$M$</td>
<td>1.07</td>
</tr>
<tr>
<td>$\Delta K_{\text{MAX}}$ (MPa)</td>
<td>$10^9$</td>
</tr>
</tbody>
</table>

When $e_m$ is less than $e_{m\text{min}}$, then the exponential approximation of the bulk modulus increment defined in Fig. A.1 was adopted. As a reference value, $\Delta K_{p,\text{MAX}}$ was assumed to be equal to $10^9$ MPa (Table A.1). This value has no physical meaning and should be understood as a contact module to prevent the penetration of particles into other particles. Introducing $\Delta K_p$ in the computations was possible thanks to the use of a self-developed numerical model, and to the implementation flexibility of the numerical software used (see Appendix D).

The increment of the microstructural strains $d\varepsilon_m$ is assumed isotropic. Its volumetric expression $d\varepsilon_{mV}$ is computed from the increment of $e_m$:

$$d\varepsilon_{mV} = \frac{-de_m}{1 + e}$$  \hfill (A.10)

where $e_m$ is obtained from the state surface represented in Fig. 2.
APPENDIX B. GEOCHEMICAL MODEL

The computation of $e_m$ needs the value of $s_{mS}$, which is linked to the relationship between the chemical potentials of microstructural and macrostructural water, $\mu_m$ and $\mu_M$. This work assumes equilibrium between water in both structural levels, and consequently their chemical potentials are assumed to be equal. According to Edlefsen & Anderson (1943), $\mu_M$ can be computed as:

$$\mu_M = \mu_{VO}(T) - \frac{WMM}{\rho_W} (s_M + s_{MO})$$

(B.1)

where $\mu_{VO}(T)$ is the chemical potential of pure water at the absolute temperature $T$, $WMM$ is the molar mass of water, and $\rho_W$ is the density of free water. As it was assumed by Truesdell & Jones (1974) and Suarez (2005), the activity $a_M$ of free (macrostructural) water for concentrations lower than 1 M can be approximated as (Garrels & Christ, 1965):

$$a_M = 1 - 0.017 \sum c_{i,M}$$

(B.2)

where $c_{i,M}$ are the molalities of dissolved anions, cations and neutral species in the macrostructural solution. A simplified geochemistry configuration was adopted, in which, together with the clay mineral, only calcium, sodium and chloride ions were considered (Navarro et al., 2017c). Electro-kinetic processes were not taken into account, and it was assumed that no significant pH changes occur. For this system, the molalities needed in Eq. (B.2) are $c_{Cl,M}$, $c_{Na,M}$ and $c_{Ca,M}$. Therefore, $s_{MO}$ is given by:

$$s_{MO} = -\frac{\rho_W}{WMM} RT Ln \left(1 - 0.017 \left(c_{Cl,M} + c_{Na,M} + c_{Ca,M}\right)\right)$$

(B.3)

where $R$ is the universal gas constant. In turn, the chemical potential of the microstructural water can be expressed as (Karnland et al., 2005):
The concentration of non charge compensating ions, which defines the value of $\Delta s_{m\text{NCCI}}$, is smaller than the concentration of ions in charge compensating positions, which affects $\mu_m$ implicitly through $p$ and $s_{mS}$. For this reason, in the previous equation, $\Delta s_{m\text{NCCI}}$ is not as relevant as $p$ and $s_{mS}$. The value of $\Delta s_{m\text{NCCI}}$ could be approximated using activity coefficients equal to 1. Nevertheless, analogously to Eq. (B.3), it was computed as:

$$
\Delta s_{m\text{NCCI}} = -\frac{\rho_m}{WMM} RT \ln \left( 1 - 0.017 \left( c_{Cl,m} + c_{Na,m\text{NCCI}} + c_{Ca,m\text{NCCI}} \right) \right)
$$

(B.5)

where $c_{Cl,m}$ is the molality of chloride in the microstructure, and $c_{Na,m\text{NCCI}}$ and $c_{Ca,m\text{NCCI}}$ are the microstructural non charge compensating molal concentration of sodium and calcium, respectively. All chloride in the microstructure will be non compensating. Anion exclusion is not imposed, and the concentration of chloride in the microstructure will be given by chemical equilibrium.

In Eqs. (B.4) and (B.5), $\rho_m$ is the density of adsorbed water. Although several studies note that $\rho_m$ is greater than the density of free water $\rho_W$ (Jacinto et al., 2012; Villar & Lloret, 2004), $\rho_m = \rho_W$ is usually assumed for modelling purposes (Tournassat & Appelo, 2011). The latter has been assumed in nine of the ten simulations performed in the present work (Table 2). On the other hand, simulation T2 uses a logarithmic function:

$$
\rho_m = \text{Max}(0.0225 \ln(s_{mS}) + 1.0545; 1)
$$

(B.6)

where $s_{mS}$ is expressed in MPa. This function is based on the values of $\rho_m$ identified by Jacinto et al. (2012) (Fig. B.1).

As deduced from Eqs. (B.1) and (B.4), in equilibrium ($\mu_m = \mu_M$), $s_{mS}$ can be calculated as in Eq. (3) ($s_{mS} = p + s_M + s_{MO} - \Delta s_{m\text{NCCI}}$), and then $\epsilon_m$ can be obtained with Fig. 2. The values of $s_{MO}$...
\( \Delta s_{mNCCI} \) can be computed with Eqs. (B.3) and (B.5), respectively. For this purpose, the concentrations of calcium, sodium and chloride in both the macrostructure and the microstructure are needed. To determine these concentrations, the Donnan equilibrium approach (Helfferich, 1962) was used. Assuming the electrochemical potential of ions are equal in both structural levels (see Navarro et al., 2017c, for details), the macrostructural and microstructural concentration of each species \((C_{i,M} \text{ and } C_{i,m}, \text{ with } i=\text{Cl, Na and Ca})\) can be related using the partition function \(B\):

\[
C_{i,M} = \frac{C_{i,m}}{B_{z_i}} \quad \text{(B.7)}
\]

where \(z_i\) is the ionic charge. Additionally, the electroneutrality of the system must be satisfied:

\[
C_{Na,M} + 2C_{Ca,M} - C_{Cl,M} = 0 \quad \text{(B.8)}
\]

\[
C_{Na,m} + 2C_{Ca,m} - C_{Cl,m} - q = 0 \quad \text{(B.9)}
\]

where \(q\) is the surface charge (mol\(_c\)/L), which can be calculated as follows:

\[
q = \frac{CEC \rho_{\text{mineral}}}{\epsilon_m} \quad \text{(B.10)}
\]

The values of \(CEC\) and bentonite mineral density \(\rho_{\text{mineral}}\) are included in Table 1. \(C_{Ca,M}\) and \(C_{Cl,m}\) are adopted as state variables of the electrochemical problem (see the “Conceptual and Numerical Model” section). By combining Eqs. (B.7) to (B.9), the following equation is obtained:

\[
-(2C_{Ca,M} + C_{Cl,m})B^2 + 2C_{Ca,M}B + C_{Cl,M} + q = 0 \quad \text{(B.11)}
\]

which is a quadratic functional expression of \(B\). \(C_{Ca,M}\) and \(C_{Cl,m}\), \(C_{Ca,m}\) and \(C_{Cl,M}\) are obtained from \(B\) by applying Eq. (B.7). Then, \(C_{Na,M}\) and \(C_{Na,m}\) can be obtained considering electroneutrality, as shown in Eqs. (B.8) and (B.9).
APPENDIX C. FLOW AND TRANSPORT FORMULATION

Since isothermal conditions and a constant gas pressure equal to the atmospheric pressure were assumed, only water and vapour flows were computed. The water mass balance equation in the macrostructure is:

$$\frac{\partial (m_M + m_V)}{\partial t} + \nabla \cdot (m_M \mathbf{v}_M + j_V) + r_m = 0$$  \hspace{1cm} (C.1)

where $m_M$ and $m_V$ are the mass of the macrostructural liquid water and vapour water per unit volume, respectively, $m_M \mathbf{v}_M$ is the mass flow of the macrostructural liquid water per unit area, $j_V$ is the mass flow of vapour, and $r_m$ is the mass exchange from macrostructural water to microstructural water. Osmotic flow terms have not been considered. The values of $m_M$ and $m_V$ are computed as:

$$m_M = \rho_w \frac{S_{rM} e_M}{1 + e}$$

$$m_V = \rho_v (1 - S_{rM}) e_M$$  \hspace{1cm} (C.2)

where $\rho_v$ is the vapour density and $S_{rM}$ is the macrostructural degree of saturation, which was calculated from the macrostructural suction (Navarro et al., 2015) using a van Genuchten (1980) retention curve:

$$\frac{S_{rM} - S_{rM,0}}{1 - S_{rM,0}} = \left[1 + (\alpha s)^{m}\right]^{-n}$$  \hspace{1cm} (C.3)

where $S_{rM,0}$ is the macropore residual degree of saturation (which is assumed to be equal to 0) and $\alpha$, $m$ and $n$ are model parameters (Table C.1). The relationship $n = 1/(1-m)$ was adopted.
Table C.1 Hydraulic parameters of the macrostructure (taken from Navarro et al., 2016).

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$ (kPa$^{-1}$)</td>
<td>$1.15 \times 10^{-4}$</td>
</tr>
<tr>
<td>$m$</td>
<td>0.733</td>
</tr>
<tr>
<td>$b_M$</td>
<td>9.911</td>
</tr>
<tr>
<td>$\phi_{M0}$</td>
<td>0.047</td>
</tr>
<tr>
<td>$k_0$ (m$^2$)</td>
<td>$2.39 \times 10^{-21}$</td>
</tr>
</tbody>
</table>

The vapour density $\rho_V$ was calculated as (Edlefsen & Anderson, 1943):

$$
\rho_V = \rho_{VO} \exp \left( -\frac{WMM (s_M + s_{MO})}{\rho_w RT} \right)
$$

(C.4)

where $\rho_{VO}$ is the density of saturated water vapour and is defined as (Ewen & Thomas, 1989):

$$
\rho_{VO} = \frac{\exp \left( 6.374 \times 10^{-2} T^\ast - 1.634 \times 10^{-4} (T^\ast)^2 \right)}{194.4}
$$

(C.5)

where $T^\ast$ is temperature expressed in degrees Celsius.

The macrostructural water flow was modelled using the conventional advective (Darcy) formulation proposed by Pollock (1986). Therefore, $v_M = v + q_M$, where $v_M$ is the macrostructural flow vector and $v$ is the time derivative of the soil skeleton displacement vector $u$. The macrostructural water seepage $q_M$ is calculated as a function of the gradients of the liquid pressure $P_L$ and the vertical coordinate $z$:

$$
q_M = -\frac{K_M \kappa_M}{\mu_w} \left( \nabla P_L + g \rho_w \nabla z \right)
$$

(C.6)

where $K_M$ is the intrinsic permeability of the macrostructural liquid, $\kappa_M$ is the macrostructural relative permeability, and $\mu_w$ is the macrostructural liquid dynamic viscosity. The relative permeability was modelled using the Brooks & Corey (1964) formulation, in which an
exponential value of three was assumed (Gens et al., 2011). The intrinsic permeability $K_M$ was modelled using the relationship proposed by Gens et al. (2011):

$$K_M = k_0 \exp\left[b_M (\phi_M - \phi_{M0})\right]$$  \hspace{2cm} (C.7)

where $k_0$ is the intrinsic permeability for the reference macrostructural porosity $\phi_{M0}$, $\phi_M$ is the macrostructural porosity, and $b_M$ is a constitutive parameter (refer to Table C.1).

The mass flow of vapour $j_V$ was considered to be diffusive only, as a constant gas pressure was assumed. Therefore, a constant gas density can be considered. In accordance with Pollock (1986):

$$j_V = -\phi_M (1-Sr_M) \tau D \nabla \rho_V$$  \hspace{2cm} (C.8)

where $\tau$ is the soil tortuosity and $D$ is the binary diffusion coefficient of water vapour in the gas. As in Olivella & Gens (2000), $\tau = 1$ was assumed, and $D$ is calculated as proposed by Pollock (1986) and Philip & De Vries (1957):

$$D = 5.9 \times 10^{-9} \frac{T^{2.3}}{P_G}$$  \hspace{2cm} (C.9)

The gas pressure $P_G$, equal to the atmospheric pressure in this study, must be expressed in kPa in this dimensional equation to obtain $D$ in m$^2$/s.

The mass exchange from macrostructural water to microstructural water $r_m$ can be deduced from the microstructural water mass balance equation. The microstructure was considered fully saturated (Yong, 1999; Mašín & Khalili, 2016), and the microstructural water was considered to be primarily linked to the bentonite skeleton. Thus, the microstructural flow vector $v_m$ was assumed to be equal to $v$, and the microstructural water mass balance equation is:

$$\frac{\partial m_m}{\partial t} + \nabla \cdot (m_m v) - r_m = 0$$  \hspace{2cm} (C.10)
where the mass of microstructural water per unit volume $m_m$ is defined as:

$$m_m = \frac{\rho_w e_m}{1 + e} \quad (C.11)$$

where $e_m$ is obtained from Eq. (3) (equilibrium between microstructural water and macrostructural water is assumed) and Fig. 2, and both $v$ and $e$ are obtained by solving the mechanical problem. Consequently, $r_m$ can be calculated as:

$$r_m = \frac{\partial m_m}{\partial t} + \nabla \cdot (m_m v) \quad (C.12)$$

The total mass balance of the species dissolved in water was determined as:

$$\frac{\partial m_i}{\partial t} + \nabla \cdot (m_i v_i) = 0 \quad (C.13)$$

where $m_i$ is the mass of the i-th ion (Ca$^{2+}$ or Cl$^{-}$ in this paper) per unit volume, which is calculated as:

$$m_i = m_{i,M} + m_{i,m} = \frac{C_{i,M} e_{M} + C_{i,m} e_m}{1 + e} \quad (C.14)$$

where $m_{i,M}$ and $m_{i,m}$ are the mass of the solute per unit volume in the macrostructure and microstructure, respectively. The term $m_i v_i$ in Eq. (C.13) is the mass flow of solute per unit area and is calculated as:

$$m_i v_i = m_i v + l_i \quad (C.15)$$

where $l_i$ is the mass flow of the i-th ion with regard to the soil skeleton displacement. This work does not consider electro-osmotic phenomena, so $l_i$ was computed as:

$$l_i = c_{i,M} q_M + j_{i,M} + j_{i,m} \quad (C.16)$$

where $c_{i,M} q_M$ defines the advective flow in the macrostructure, $j_{i,M}$ is the diffusion of the i-th ion in the macrostructure with respect to the advective flow, and $j_{i,m}$ is the diffusion of the i-th ion in the microstructural liquid. As previously noted, water flow in the microstructure was
not considered. In addition, mechanical dispersion was neglected because the advective flow was small. Therefore, diffusion was modelled using Fick's law:

\[
\begin{align*}
\mathbf{j}_{\mathrm{M}} &= -\frac{e\rho}{1 + e} D_{\mathrm{M}} \nabla C_{\mathrm{M}} \\
\mathbf{j}_{\mathrm{m}} &= -\frac{e\rho}{1 + e} D_{\mathrm{m}} \nabla C_{\mathrm{m}}
\end{align*}
\]  

(C.17)

where \( D_{\mathrm{M}} \) and \( D_{\mathrm{m}} \) are the macrostructural molecular diffusion coefficient and microstructural molecular diffusion coefficient of the i-th ion, respectively. To determine \( D_{\mathrm{M}} \) and \( D_{\mathrm{m}} \), the formulation of Bourg et al. (2006) is used:

\[
\begin{align*}
D_{\mathrm{M}} &= D_{\mathrm{o,i}} \frac{1}{G} \\
D_{\mathrm{m}} &= D_{\mathrm{o,i}} \frac{\delta}{G}
\end{align*}
\]  

(C.18)

where \( D_{\mathrm{o,i}} \) is the self-diffusion coefficient of the i-th ion in bulk liquid water, \( G = 4 \) is a factor of the pore-network geometry, and \( \delta = 0.3 \) is a constrictivity factor for small pores. It is possible that the value of these two factors, especially \( G \), changes when bentonite deforms and its particles are rearranged. However, constant values were taken based on the estimations by Bourg et al. (2006) for bentonites similar to the one considered in the present work. Soil strains are already accounted for in Eq. (C.17) through void ratios. The values taken for the self-diffusion coefficients are \( D_{\mathrm{o,\text{Ca}}} = 7.79 \times 10^{-10} \) m\(^2\)/s and \( D_{\mathrm{o,\text{Cl}}} = 2.03 \times 10^{-9} \) m\(^2\)/s (Cussler, 1997). For case T1 (Table 2), changes in the values of \( D \) (\( D_{\mathrm{i,M}} \) and \( D_{\mathrm{i,m}} \), with i=Ca and Cl) up to a 20% have been confirmed to produce changes in the predicted extract salinity lower than 5.5%. Consequently, the sensitivity of the performed analysis to the definition of molecular diffusion is limited.
APPENDIX D. NUMERICAL IMPLEMENTATION

Eqs. (C.1), (C.13) and (A.2) define the nonlinear system of coupled partial differential equations that must be solved to obtain $P_L$, $C_{Ca,M}$, $C_{Cl,m}$ and $u$, state variables of the system. Given the multiphysics character of the problem, the use of a multiphysics partial differential equation solver is very convenient. In such solvers, the software automatically manages discretisation and numerically processes the differential equations that describe the analysed physical phenomena, on which the user can focus. Some of the solvers, like Comsol Multiphysics (COMSOL, 2015), use a symbolic algebra interface. The equations to be solved, and the functions involved, can be introduced in a text-editor-like interface. The system is flexible in the definition the equations to be solved, and their implementation is simple. Comsol Multiphysics also offers prebuilt modules to solve mechanical equilibrium or parabolic equations. However, these modules are not as flexible as the “blank” equations available. The authors of this work implemented their own modules from blank equations, and included the equations described in the previous appendices.

In addition, the symbolic algebra performs automatic differentiation, which improves the numerical performance of the solver (see, for instance, Bischof et al., 2008; Martins & Hwang, 2012). However, the use of a symbolic iteration matrix can cause problems when there are state functions defined implicitly. This is the case, for instance, of the mechanical constitutive model, since the increments of stress, strain and plastic variables depend on the value of the variables themselves. The problem can be overcome using a mixed method (Malkus & Hughes, 1978), that is, defining stresses and plastic variables as state variables (Navarro et al., 2014). The application of this method has been confirmed efficient and to produce quality results (Alonso et al., 2012; Navarro et al., 2014).
## NOTATION

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$B$</td>
<td>Partition coefficient for ion concentrations in the macrostructure and the microstructure</td>
</tr>
<tr>
<td>$CEC$</td>
<td>Cation exchange capacity</td>
</tr>
<tr>
<td>$C_{i,M}$</td>
<td>Molar concentration of the $i$-th (Cl$^-$, Na$^+$ and Ca$^{2+}$) in the macrostructural water</td>
</tr>
<tr>
<td>$C_{i,m}$</td>
<td>Molar concentration of the $i$-th (Cl$^-$, Na$^+$ and Ca$^{2+}$) in the microstructure</td>
</tr>
<tr>
<td>$c$</td>
<td>Salinity of the macrostructural solution</td>
</tr>
<tr>
<td>$e$</td>
<td>Total void ratio</td>
</tr>
<tr>
<td>$e_M$</td>
<td>Macrostructural void ratio</td>
</tr>
<tr>
<td>$e_{M\text{min}}$</td>
<td>Value of the macrostructural void ratio in which the definition of the macrostructural stress-strain behaviour changes</td>
</tr>
<tr>
<td>$e_m$</td>
<td>Microstructural void ratio</td>
</tr>
<tr>
<td>$e_{mR}$</td>
<td>Remaining microstructural void ratio under dry conditions</td>
</tr>
<tr>
<td>$IS$</td>
<td>Ionic strength</td>
</tr>
<tr>
<td>$l_i$</td>
<td>Mass flow of the $i$-th ion with regard to the soil skeleton displacement</td>
</tr>
<tr>
<td>$P_{atm}$</td>
<td>Atmospheric pressure</td>
</tr>
<tr>
<td>$P_G$</td>
<td>Gas pressure</td>
</tr>
<tr>
<td>$P_L$</td>
<td>Macrostructural liquid pressure</td>
</tr>
<tr>
<td>$p$</td>
<td>Net mean stress</td>
</tr>
<tr>
<td>$p'$</td>
<td>Effective mean stress</td>
</tr>
<tr>
<td>$q_M$</td>
<td>Macrostructural water seepage</td>
</tr>
<tr>
<td>$s_M$</td>
<td>Macrostructural matric suction</td>
</tr>
<tr>
<td>$s_{MO}$</td>
<td>Macrostructural osmotic suction</td>
</tr>
<tr>
<td>$s_{mS}$</td>
<td>Structural suction of the microstructure</td>
</tr>
<tr>
<td>$S_{rM}$</td>
<td>Macrostructural degree of saturation</td>
</tr>
<tr>
<td>$t_{\Delta \sigma}$</td>
<td>Time to reach the maximum squeezing pressure applied</td>
</tr>
<tr>
<td>$u$</td>
<td>Soil skeleton displacement vector</td>
</tr>
<tr>
<td>$w$</td>
<td>Total gravimetric water content</td>
</tr>
<tr>
<td>$\Delta s_{mNCCI}$</td>
<td>Osmotic pressure caused by the non charge compensating ions in the microstructure</td>
</tr>
<tr>
<td>$\Delta \sigma$</td>
<td>Squeezing pressure</td>
</tr>
</tbody>
</table>
\( \rho_0 \)  Dry density
\( \rho_m \)  Density of adsorbed water
\( \rho_{\text{mineral}} \)  Mineral density
\( \sigma_R \)  Total radial stress
\( \sigma_Z \)  Total vertical stress

REFERENCES


Table 1. Main properties of the tested material, where CEC is the cation exchange capacity and $\rho_{\text{mineral}}$ is the bentonite density.

<table>
<thead>
<tr>
<th>Chemical composition (weight %)</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO$_2$</td>
<td>59.82</td>
</tr>
<tr>
<td>Al$_2$O$_3$</td>
<td>21.27</td>
</tr>
<tr>
<td>Fe$_2$O$_3$</td>
<td>3.62</td>
</tr>
<tr>
<td>Na$_2$O</td>
<td>2.86</td>
</tr>
<tr>
<td>MgO</td>
<td>2.77</td>
</tr>
<tr>
<td>CaO</td>
<td>1.49</td>
</tr>
<tr>
<td>Mineralogical composition (weight %)</td>
<td></td>
</tr>
<tr>
<td>Smectite</td>
<td>87.6</td>
</tr>
<tr>
<td>Plagioclase</td>
<td>4.2</td>
</tr>
<tr>
<td>Quartz</td>
<td>4.1</td>
</tr>
<tr>
<td>K-feldspar</td>
<td>1.8</td>
</tr>
<tr>
<td>Other properties</td>
<td></td>
</tr>
<tr>
<td>$CEC$ (eq/kg)</td>
<td>0.84</td>
</tr>
<tr>
<td>$Na^+$/K$^+$/Ca$^{2+}$/Mg$^{2+}$ (eq/kg)</td>
<td>0.58/0.02/0.25/0.08</td>
</tr>
<tr>
<td>$\rho_{\text{mineral}}$ (g/cm$^3$)</td>
<td>2.78</td>
</tr>
<tr>
<td>Liquid % / Plastic Limit %</td>
<td>510/50</td>
</tr>
</tbody>
</table>

Table 2. Description of the simulated squeezing tests.

<table>
<thead>
<tr>
<th>ID</th>
<th>$\rho_D$ (g/cm$^3$)</th>
<th>$\rho_m$ (g/cm$^3$)</th>
<th>Water</th>
<th>$c$ (g/L)</th>
<th>$\Delta\sigma$ (MPa)</th>
<th>$t_{\Delta\sigma}$ (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>T1</td>
<td>0.7</td>
<td>1</td>
<td>SAT</td>
<td>10</td>
<td>40</td>
<td>5</td>
</tr>
<tr>
<td>T1a1</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>T1a2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>T1b1</td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>T1b2</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>T1c1</td>
<td></td>
<td></td>
<td></td>
<td>1</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>T1c2</td>
<td></td>
<td></td>
<td></td>
<td>35</td>
<td>80</td>
<td>5</td>
</tr>
<tr>
<td>T2</td>
<td>Eq. (B.6)</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>T3</td>
<td>1.4</td>
<td>1</td>
<td></td>
<td>10</td>
<td>40</td>
<td>5</td>
</tr>
<tr>
<td>T4</td>
<td>1.4</td>
<td>1</td>
<td>$w=17%$</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$w=17\%$
List of figure captions

Fig. 1. (a) Mechanical boundary conditions of the simulated squeezing tests. (b) Idem for flow and transport. (c) Reference points, A: \( z=0 \), B: \( z=0.1 \) \( H \), C: \( z=0.5 \) \( H \).

Fig. 2. State surface (line) that defines the microstructural volumetric constitutive model and the experimental data (markers) that were used to deduce the model.

Fig. 3. (a) Water content distributions at the end of the tests performed by Navarro et al. (2017c) (dots are experimental data, and lines are model predictions). (b) Correlation between the experimental and model results of the tests of Karnland et al. (2005, 2006) (salinities of 0.1, 0.3 and 1 M were considered).

Fig. 4. Evolution of the mean molar concentrations of the ions in the extract and the initial values for the macrostructure. (a) Tests T1, T1a1 and T1a2. (b) Tests T1, T1b1 and T1b2. See Table 2.

Fig. 5. Evolution of the mean molar concentrations of the ions in the extract and the initial values for the macrostructure of tests T1c1 (a), T1 (b) and T1c2 (c). See Table 2.

Fig. 6. Evolution of the total vertical stress \( \sigma_Z \) (equal to the applied load), \( P_L \), \( p' \), \( s_m \), and \( s_{MO} - \Delta s_{mNCCI} \) of test T1. (a) Point A, (b) Point B, (c) Point C. See Table 2 and Fig. 1(c).

Fig. 7. Evolution of \( e \), \( e_M \) and \( e_m \) at points A, B and C (Fig. 1 c). (a) Test T1b1. (b) Test T1. (c) Test T1b2. See Table 2.

Fig. 8. Volumes of macrostructural water and microstructural water extracted. a) Test T1b1. (b) Test T1. (c) Test T1b2. See Table 2.

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Fig. 12. Percentage variation in the number of ions considered in the model. Test T1 (Table 2).

Fig. 13. Evolution of the mean molar concentrations of the ions in the extract and the initial values for the macrostructure in Test T3 (Table 2).

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Fig. 15. Evolution of $e$, $e_M$ and $e_m$ at point A shown in Fig. 1 throughout Test T4 (Table 2).

Fig. 16. Evolution of the total vertical stress $\sigma_Z$, total radial stress $\sigma_R$, $P_L$, $p$, $s_m$, and $s_{MO} - \Delta s_{mNCI}$ in Test T4 (Table 2) at point A shown in Fig. 1(c).

Fig. A.1. Increase $\Delta K_p$ in the bulk modulus $K_p$ computed when $e_M$ is less than $e_{M\min}$.

Fig. B.1. Value of $\rho_m$ with respect to $s_{mS}$ (adapted from Jacinto et al., 2012).
Navarro Fig. 01

(a) Squeezing pressure

(b) No normal displacement

(c) No flow

Eq. (1)
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