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AN ELASTOPLASTIC MODEL OF BENTONITE FREE SWELLING

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ABBREVIATIONS

BBM

BExM

FS

HLW

Barcelona Basic Model

Barcelona Expansive Model

Free Swelling

High Level radioactive Waste

ABSTRACT

This article proposes a new plastification mechanism to model bentonite free swelling processes by using an elastoplastic approach. The formulation is based on an interaction function that defines the plastic macrostructural strains induced by microstructural effects. This term is defined as a function of the microstructural void ratio, and it establishes a direct connection between the destructure of the micro- and macrostructures of the bentonite during the free swelling processes. The formulation proposed also includes a new approach to the calculation of the microstructural void ratio as well as a new method to describe the kinetic mass exchange between micro- and macrostructural water, considering that there may not be equilibrium between the two types of water. The model was used to analyse several free swelling tests and was found to yield satisfactory results.

KEYWORDS

Expansive soils; Bentonite free swelling; Plasticity; Partial saturation; Soil fabric/structure

1. INTRODUCTION

Because of the physical and chemical properties of bentonite clays, they have been considered in several countries for use as buffer and backfill materials in spent nuclear fuel repositories. Specifically, in the KBS-3 disposal concept (developed by the Swedish nuclear fuel and waste management administration, SKB, 1999, and by Posiva, 2006, the Finnish expert organisation responsible for the spent nuclear fuel repository), dry Na-bentonite of the Wyoming type (called “MX-80”; see Karnland et al., 2005, and Dueck & Nilsson, 2010) is considered for placement around high level radioactive waste (HLW) canisters in a highly compacted state. A reliable model of the behaviour of these bentonites is needed to assess the safety performance of the barrier. For this purpose, the model should characterise the bentonite free swelling (FS) associated with erosion (see, for instance, Birgersson et al., 2009, and Moreno et al., 2010), a process of special interest in the field of designing HLW repositories.

Some authors have proposed sol-gel transition models to describe the soil behaviour close to this extreme situation (Liu et al., 2009). However, despite their value, at the present time these models are not able to describe the entire swelling process of bentonite, from an initial unsaturated (with values of suction on the order of tens to hundreds of MPa) and well structured (both micro and macrostructurally) state to the release of soil particles when it becomes a sol.

This study proposes an elastoplastic formulation that contributes to the characterisation of the saturated and unsaturated FS of bentonites. First, a review of its basic concepts will be presented. Afterwards, an analysis of its application to the simulation of FS is illustrated by several examples.

2. THEORY (1): BASIS

Since the seminal work of Collins & McGown (1974) and Collins (1984), a number of researchers have observed the existence of two structural levels in compacted clays (Lloret et al., 2003; Pusch, 1982; Pusch & Moreno, 2001; Romero et al., 1999; Romero & Simms, 2008). Thus, the structure of the bentonites analysed in this paper is idealised into two structural levels: macro and micro structures. The microstructural level is associated with both intersheet voids and intra-aggregate pores (see Fig. 1 a, adapted from Pusch, 1987). The pores between the aggregates, inter-aggregate pores or macropores (Romero et al., 2011), define the macrostructural level.

Like the formulations derived from Gens & Alonso (1992), the one presented here foregoes the characterisation of the internal topology of the aggregates. Although it is reasonable to assume that at least two different hierarchical levels exist, interlamellar voids and interstack voids (following the notation in Neretnieks et al., 2009), only one macroscopic variable, the microstructural void ratio e_m (e_m = volume of voids in the microstructure per volume of mineral), has been considered for characterising the intra-aggregate porosity (microporosity). The aggregate has been adopted as the support scale (according to Pachepsky et al., 2006) of the microstructure, performing an abstraction of its internal complexity.

In accordance with Gens & Alonso (1992), the microstructural void ratio variation is assumed to be associated with the variation of the intra-aggregate water content.

According also to these authors and to Hueckel (1992) and Quirk (1994), the microstructure deformation is assumed to be fully reversible and unaffected by the macrostructure deformation. However, when considering the deformation of the aggregates, it must be taken into account that their swelling (micro-swelling) can occlude the macropores to different extents (Mašín, 2013). Given their random arrangement (Quirk, 1994), each aggregate (micro-domain) swells in a different

direction, and micro deformations induce macro deformations (Cui et al., 2002; Khalili et al., 2010; Villar & Lloret, 2001). This coupling causes a complex array of behavioural trends (Della Vecchia et al., 2013; Koliji et al., 2010; Sun & Sun, 2012), which are adequately described by the “Barcelona Expansive Model” (BExM; Alonso et al., 1999) for compacted bentonites (see Lloret et al., 2003). To achieve this description, the BExM introduces two interaction functions (Alonso et al., 1999): f_C , when microstructural compression is produced (i.e., e_m decreases), and f_S for microstructural swelling (i.e., e_m increases). The shape of these interaction functions is depicted in Fig. 2. The value of f_C and f_S depends on the degree of openness of the macrostructure relative to the applied stress state (Sánchez et al., 2005). The degree of openness is determined by the p/p_O ratio in isotropic stress states, where p is the net mean stress ($p=p_{TOT}-P_G$, where p_{TOT} is the mean stress and P_G is the gas pressure) and p_O is the net mean yield stress at the current suction (see Fig. 3). If a deviatoric stress exists, the openness is determined by p_R/p_O (Sánchez et al., 2005), where the reference stress p_R is defined in Fig. 3. Once f_α is defined (α = “C” if $de_m < 0$ and α = “S” if $de_m > 0$, where de_m is the increment of the microstructural void ratio), the macrostructural plastic volumetric strains induced by microstructural effects, $d\varepsilon_{MV,m}^P$, can be expressed as:

$$[1] \quad d\varepsilon_{MV,m}^P = f_\alpha \cdot d\varepsilon_{mV}$$

where $d\varepsilon_{mV}$ is the microstructural volumetric strain, which is supposed to be elastic.

Therefore, it is assumed that irreversible macrostructural plastic strains are induced whenever microstructural deformations occur.

Functions f_α are constitutive terms. To define them, Alonso et al. (1999), Sánchez et al. (2005) and Gens et al. (2011) considered not only different parameters but also different “shapes” for different soils. Different ways of defining $d\varepsilon_{mV}$ have been considered (see, for instance, Mašín, 2013; Musso et al., 2013; Romero et al., 2011). Sánchez et al.

(2005) take into account both exponential and logarithmic laws to define $d\varepsilon_{mV}$ in terms of the variation of the microstructural effective stress. In this paper, e_m is calculated by a one-to-one model based on the formulation by Zhang et al. (1995):

$$[2] \quad \frac{1}{e_m} = \frac{1}{e_{mO}} + \frac{1}{k_m} \cdot \text{Ln} \left(1 + \frac{\pi}{P_{\text{ATM}}} \right)$$

where k_m is a micro deformability parameter and e_{mO} is the microstructural void ratio when the bentonite swelling pressure π is equal to 0, that is, when only the atmospheric pressure P_{ATM} is applied. Note that the swelling pressure is interpreted as the pressure exerted by the aggregates when swelling occurs in pure water, that is, when the vapour pressure is set only by the clay skeleton.

The analyses used by Zhang et al. (1995) to derive Eq. (2) were conducted with bentonites in which the void ratio reached high values (greater than 1). This formulation is valuable when simulating free swelling processes. However, this casts doubt on the validity of Eq. (2) for compacted bentonites in which the void ratio may be substantially lower. This issue has recently been analysed by the authors of the present study (Navarro et al., 2013). Their results are synthesised in Fig. 4, where the model and experimental results of the swelling of individual bentonite aggregates are compared. The experimental results were obtained by Montes-H et al. (2003) through the combined application of environmental scanning electron microscopy and digital image analysis. The satisfactory fits in Fig. 4 provide confidence in Eq. (2) for a wide range of values of the microstructural void ratio.

The model results depicted in Fig. 4 were obtained considering that there may not be equilibrium between microstructural and macrostructural water. Therefore, water mass exchange c_m between the two types of water was taken into account (Navarro et al., 2013). As in other phase change processes (de Groot & Mazur, 1984), c_m may be

assumed to depend on the difference between the chemical potential of the (microstructural) water inside the aggregates, μ_m , and the chemical potential of the macrostructural water, μ_M . In low salinity conditions, the microstructural water potential may be calculated by (Karnland et al., 2005):

$$[3] \quad \mu_m = \mu_{VO}(T) + \frac{WMM}{\rho_w} \cdot (p - \pi)$$

where $\mu_{VO}(T)$ is the reference chemical potential of water vapour over a flat surface of pure water at absolute temperature T , WMM is the water molar mass and ρ_w is the microstructural water density, which is assumed to be equal to that of free water (Tournassat & Appelo, 2011). The chemical potential of the macrostructural water can be defined, also in diluted conditions, as (Edlefsen & Anderson, 1943):

$$[4] \quad \mu_M = \mu_{VO}(T) - \frac{WMM}{\rho_w} \cdot s$$

where s is the macrostructural water matric suction ($s = P_G - P_L$, P_L being the pressure of the macrostructural liquid). Thus, according to Eqs. (3) and (4), $\mu_M - \mu_m$ may be calculated as:

$$[5] \quad \mu_M - \mu_m = \frac{WMM}{\rho_w} \cdot (\pi - (p + s))$$

If π_B is defined as:

$$[6] \quad \pi_B = p + s$$

the difference $\pi - \pi_B$ may be understood as the “thermodynamic force” which induces the flow c_m . In this sense, π_B has the meaning of a “boundary swelling pressure” (Navarro et al., 2008). Although the concept is valid even for moderately to highly saline solutions, the boundary swelling pressure definition given in Eq. (6) will become more complicated for those conditions, as the activities of the solutes

dissolved in the macro and microstructural water have to be taken into account in Eqs. (4) and (5). Unlike other authors (Gerke & van Genuchten, 1993 a, 1993 b; Musso et al., 2013) that propose a first-order water transfer model, this study adopts a non-linear formulation to define the mass exchange c_m between the two types of water (Navarro et al., 2013):

$$[7] \quad \frac{c_m}{\rho_w} = H \cdot \left(\frac{\pi_B}{\pi} \right)^C \cdot (\pi - \pi_B)$$

where H and C are constitutive parameters. H defines the transfer coefficient at the end of the mass exchange process (when $\pi = \pi_B$) and C synthesises the way in which the mass transfer term changes as π approaches π_B .

Equation (7), together with Eqs. (2) and (6), were used in the simulation of the swelling tests analysed in Section 4. These three equations define an alternative approach to the approach provided by other authors (Gens et al., 2011, Alonso et al., 2011) to characterise the microstructure volumetric behaviour and the transitory processes caused by the mass exchange between macrostructural and microstructural water. However, the formulation raised so far does not introduce any means to modify the coupling between the microstructural swelling and macrostructural deformation defined by the function f_s .

3. THEORY (2): PROPOSE FREE SWELLING AS A PLASTIC MECHANISM

According to Fig. 2 and Eq. (1), the swelling (negative volumetric strains, following the Soil Mechanics signs convention) of the microstructure causes negative plastic volumetric deformations of the macrostructure. Therefore, according to the hardening law (Alonso et al., 2011):

$$[8] \quad dp_o^* = \frac{(1+e) p_o^*}{\lambda(0) - \kappa(s)} d\varepsilon_{MV}^p$$

microstructural swelling would cause a decrease in the saturated pre-consolidation stress p_O^* , that is, a macrostructural softening. In Eq. (8), e is the total void ratio (e = total volume of voids in the soil per volume of mineral), $\lambda(0)$ and $\kappa(s)$ are model functions defined in Appendix B and $d\varepsilon_{MV}^p$ is the increment of the macrostructural plastic volumetric strain.

However, if p_R/p_O is equal to 1, f_S will reach its minimum value (see Fig. 2). If that value is negative, the micropores will be assumed to invade the macropores, reducing the macrostructural porosity and giving rise to macrostructural hardening. This progressive clogging of the macrostructural porosity was suggested by Pusch & Hökmark (1990) and was observed by Komine & Ogata (1994) by using environmental scanning electron microscopy, both considering restricted deformation conditions. Nevertheless, under FS conditions, the swelling can continue. In this case, for sodium dominated clays, sheets tend to separate mostly into individual sheets (Pusch & Yong, 2006; Neretnieks et al., 2009). If bentonite gels are formed, house-of-cards-like structures in which edge-to-face interactions come into play will be formed (Pusch, 1982; Birgersson et al., 2009). This causes the destructuring of the aggregates contained in the compacted bentonite, which leads to a large increase in the volume of the macrostructure and its destructuration. This process was observed by Pusch (1987) when studying a Wyoming bentonite using a high voltage electron microscope. As shown qualitatively in Fig. 1, the percentage of microstructural porosity decreases in relative terms. While there is an increase in the macrostructural pores, their maximum size is smaller. This was also observed in a study on bentonite swelling under constant load conditions (Lloret & Villar, 2007). Ye et al. (2009), in a study of the evolution of the microstructure of a Na-bentonite subjected to suction changes under unconfined

conditions, corroborated that the macroporosity underwent a greater increase than the microporosity.

The increase in macroporosity may be pronounced, as depicted in Fig. 5. This figure was obtained by identifying the internal water as defined by Pusch et al. (1990) with the microporosity, and by assuming that the definition of the macropores given by Bourg et al. (2006) is equivalent to the one put forth in this study. With these definitions, the obtained e_m/e_M values are similar to those identified by Romero et al. (2011). In the last study, the results show that the increase of e_M with respect to e_m is not sharp. However, the results presented by Bourg et al. (2006) indicate a sharper increase of e_M with respect to e_m for higher e_m values. Moreover, the e_M/e_m slope is much steeper in the higher e_m range of the curves derived from the data by Pusch et al. (1990). This behaviour is difficult to reproduce with the shape of f_S defined in Fig. 2.

For example, assume that a conventional FS test is performed. There is an axisymmetric stress state with almost null vertical stress. At the start of the test, p_R is low. However, the progressive increase in the radial stress as a consequence of the restrained swelling in the radial direction causes p_R to rise. Moreover, because p_O is still higher than p_R , f_S leads to macrostructural softening and to the gradual decrease of p_O . As the values of p_O and p_R become closer (higher p_R/p_O ratio), f_S becomes less important (see Fig. 2).

Therefore, the macrostructural deformation rate tends to decrease. Nevertheless, if, as observed in the tests analysed in the next section, p_R and p_O become equal, macrostructural yielding will take place, thus increasing the dilatant behaviour and the softening introduced by f_S previously. This leads to an increase in soil swelling.

However, as observed in the next section, the dilatant behaviour of the bentonite macrostructure is not usually important enough to explain the type of behaviour shown in Fig. 5. Therefore the formulation of the interaction function f_S should be modified to

introduce the effect exerted by the swelling of the aggregates on the macrostructure under FS conditions.

The micro-to-macro interaction function should not only characterise the reorganisation that occurs in the macrostructure when the aggregate swelling is hindered by the packing density p/p_0 . It should also model the destructuring of the macrostructure when the aggregates become destructured under FS conditions. If the reorganisation imposed by packing and the destructuration during FS are assumed to be two different active plastic mechanisms, an additive formulation can be used (Rizzi et al., 1996; Sánchez et al., 2005):

$$[9] \quad d\varepsilon_{MV,m}^p = d\varepsilon_{MV,m1}^p + d\varepsilon_{MV,m2}^p = f_1 \cdot d\varepsilon_{mV} + f_2 \cdot d\varepsilon_{mV} = (f_1 + f_2) d\varepsilon_{mV}$$

where $d\varepsilon_{MV,m1}^p$ is the macrostructural plastic volumetric strain caused by macrostructural packing (modelled, for example, with the interaction function f_s illustrated in Fig. 2, see Section 2) and $d\varepsilon_{MV,m2}^p$ is the plastic volumetric strain induced in the macrostructure by the aggregate destructuration. Hence, from an operational point of view, free swelling is considered to be a new plastic mechanism characterised by the term f_2 . It is added to f_s , designated by f_1 in the new notation.

Because the increase in e_m marks the progressive destructuring of both the microstructure and the macrostructure, f_2 is defined in terms of e_m . As a preliminary approach, the linear law defined in Fig. 6 b is suggested. In this figure, e_{mFS} indicates the value of e_m at which the destructuring of the macro is assumed to begin, and e_{mR} is the value at which the destructuring trend is the greatest and equal to f_{2M} . Therefore, e_{mR} should be understood as a parameter of the simplified model of f_2 . This linear law is most likely an over-simplification. It should be interpreted as a preliminary approach to the trend. As can be surmised from Fig. 5, the shape of f_2 will be more complicated. In particular, it seems reasonable to expect the slope of f_2 to increase notably for high e_m

values, pointing out an increment of the velocity of the macrostructure destructuring. However, the linear model is a useful tool for analysing the extent to which the new plastic mechanism, characterised by f_2 , allows for the consistent simulation of FS processes. Similarly, a linear law was also assumed for f_1 as a preliminary approach (see Fig. 6 a).

4. RESULTS AND DISCUSSION

To study the effect of f_2 , the first step was to analyse three FS tests carried out in the laboratories of B+Tech in Helsinki, Finland. They used natural MX-80 bentonite samples that were hydrated with de-ionised water (see Sane et al., 2013). Dueck & Nilsson (2010) present a detailed description of the material. The general configuration of the tests is described in Fig. 7. The initial height of the sample was 15.85 mm in test T1, 8.80 mm in test T2 and 40.00 mm in test T3. In all three tests, the sample diameter was 50 mm, which remained constant during the tests. In tests T1 and T2, the initial gravimetric water content (10%) and initial dry density (1.6 g/cm^3) were the same. In test T3, the gravimetric water content was 17% and the initial dry density was 1.73 g/cm^3 . In keeping with Kiviranta & Kumpulainen (2011), the particle density was 2.78 g/cm^3 . The experimental results are plotted in Figs. 8 a, b and c. The data derived from these tests served as a reference to compare to the simulations.

In the simulations, the BExM was adopted as the stress-strain model for the calculations. The formulation presented by Lloret et al. (2003) was used. When the BExM is adopted, the macrostructure deformation is modelled using the “Barcelona Basic Model” (BBM; Alonso et al., 1990). The formulation by Alonso et al. (2011) has been implemented, with the exception of κ_s (see Appendix B), adopted from Hoffmann et al. (2007). This formulation was implemented in a numerical model. The

multiphysics partial differential equation solver COMSOL Multiphysics (COMSOL AB, 2011) was used for this purpose. The numerical strategies described by Alonso et al. (2012) and Navarro et al. (2014) were used. The results of Lloret et al. (2003) were used for validation.

After the validation of the program, two modifications were introduced to improve its performance in the FS simulation. First, only the effect of the interaction function f_s , and not that of f_c , was taken into account. As stated in Section 2, the swelling of the microstructure always induced macrostructural plastic strains. Second, in all the simulations presented below, Eq. (2) was used to describe the volumetric behaviour of the microstructure. Parameters $e_{m0} = 21$ and $k_m = 6.6$ were used. The definition of e_{m0} took into account the findings of Zhang et al. (1995) for a Wyoming bentonite in almost fresh water (electrolyte concentration of 10^{-4} M). The value of k_m was taken from the analysis of the tests carried out by Montes-H et al. (2003) (see Fig. 4).

After making these changes, two macro-micro interaction mechanisms were analysed. The first one, M00, does not include f_2 ($f_s = f_1$, see Figs. 2 and 6) and an instantaneous equilibrium between macro and micro water is assumed ($\mu_m = \mu_M$). On the other hand, the second mechanism, M11, takes into account the effect of f_2 ($f_s = f_2 + f_1$; Eq. (9), Figs. 2 and 6) and uses Eqs. (6) and (7) to model the mass exchange between macro and microstructural water. The values of 0.4 and $1.5 \cdot 10^{-8} \text{ (kPa} \cdot \text{s)}^{-1}$ were adopted for the parameters C and H , respectively. These values are the mean of those identified by Navarro et al. (2013) after the analysis of the results obtained by Montes-H et al. (2003) (Fig. 4).

When c_m (the macro to micro water mass exchange) was taken into account, the macrostructural water mass balance equation was:

$$[10] \quad \frac{\partial m_M}{\partial t} + \nabla \cdot (m_M \cdot \mathbf{v}_M) + c_m = 0$$

where $\partial/\partial t$ is the time derivative operator. The scalar field m_M is the mass of macrostructural water per unit volume. It is defined as $m_M = (\rho_w \cdot S_{rM} \cdot e_M) / (1+e)$, where ρ_w is the density of water, S_{rM} is the macrostructural degree of saturation and e_M is the macrostructural void ratio (volume of voids in the macrostructure per volume of mineral). The experimental water content data obtained by B+Tech in separate tests were employed to fit the van Genuchten (1980) retention curve:

$$[11] \quad \frac{S_{rM} - S_{rM,0}}{1 - S_{rM,0}} = \left[1 + (\alpha \cdot s)^n \right]^{-m}$$

After assuming a macropore residual degree of saturation $S_{rM,0}$ equal to 0, the parameters $\alpha = 3.74 \times 10^{-5} \text{ kPa}^{-1}$ and $m = 0.45$ were identified. The relationship $n = 1/(1-m)$ was assumed. The macrostructural water flow vector $\mathbf{v}_M$ was computed using a conventional advective (Darcy) (Pollock, 1986) formulation, assuming $\mathbf{v}_M = \mathbf{v}_S + \mathbf{q}_M$, where $\mathbf{v}_S$ is the soil skeleton's deformation rate and $\mathbf{q}_M$ is the macrostructural water seepage. It was calculated as:

$$[12] \quad \mathbf{q}_M = - \frac{k_M \cdot \kappa_M}{\mu_w} \cdot (\nabla P_L + \rho_w \cdot \mathbf{g} \cdot \nabla z)$$

where k_M is the isotropic intrinsic permeability of the macrostructural liquid, κ_M is the macrostructural relative permeability, ∇ is the gradient differential operator, g is the gravity constant, z is the vertical coordinate and μ_w is the dynamic viscosity of water.

The vapour flow was not considered, and isothermal conditions were assumed. The relative permeability was modelled using the Brooks & Corey (1964) and Burdine (1953) formulation with an exponent value of 3, obtained in several infiltration tests on homogeneous compacted bentonite (Gens et al., 2011). The intrinsic permeability was modelled using the relationship and mean parameters proposed by Liu et al. (2011) for a natural MX-80 bentonite in distilled water.

Moreover, when equilibrium conditions between the micro and macro water were not assumed, the microstructural water mass balance equation was:

$$[13] \quad \frac{\partial m_m}{\partial t} + \nabla \cdot (m_m \cdot \mathbf{v}_m) - c_m = 0$$

where the mass of microstructural water per unit volume m_m was defined as

$m_m = (\rho_w \cdot e_m) / (1 + e)$. The microstructural water was supposed, like the immobile water of van Genuchten & Wierenga (1976), to be mainly linked to the soil skeleton, and thus $\mathbf{v}_m = \mathbf{v}_s$ was assumed.

In addition to the simulations carried out with the macro-micro interaction mechanisms M00 and M11, the free swelling tests were also simulated using the BBM (Appendix B).

In all three simulations (M00, M11 and BBM), the mechanical parameters in Table 1 were used. With the exception of α_{sp} , these parameters were obtained by B+Tech in tests independent from those analysed here. According to B+Tech tests, α_{sp} should be assigned a value of -0.145. Nevertheless, a better fit was obtained using a value of 0, which is consistent with the values identified by several authors (Chen & Ledesma, 2009; Jacinto, 2010; Kristensson & Akesson, 2008) for other MX-80 bentonites.

In both M00 and M11 simulations, f_{1M} was assigned a value of 1, which is similar to that used by Sánchez et al. (2005) and Gens et al. (2011) for different clays. In the M11 simulation, f_{2M} was considered to be equal to f_{1M} as a preliminary approach. Moreover, the maximum void ratio described in Eq. (2) was assumed to have a value close to that representing the highest destructuring capacity of the microstructure. Hence (see Fig. 6), $e_{mR} = e_{mO}$ was assumed. Besides, $e_{mFS} = 2.7$ was adopted. This value was obtained by assuming a sheet thickness of 1 nm and accounting for the change that occurs in the

distribution of the microstructural water when the average separation between sheets is equal to 2.7 nm (Low, 1987).

Experimental and model results were compared in Figs. 8 a, b and c. It is important to note that, although many of the parameters used are “tentative” (that is, they were not obtained through direct experimentation with the modelled bentonites), the fit of the simulations is satisfactory. Although these fits must be interpreted with caution, they indicate that the parameters used are feasible and that it is reasonable to derive conclusions from the comparison of the results of the models.

Figs. 8 a, b and c show that the results obtained with BBM are improved by the mechanisms that explicitly introduce the expansive character of bentonite, as expected. After observing Fig. 8 a, the results of M11 do not seem to offer any advantage with regard to those obtained with M00. However, differences are shown in Figs. 8 b and c. These figures reveal the sensitivity of the results obtained with M00 to the size of the sample. If the sample is shorter (Fig. 8 b), its average water content increases faster. For this reason, by assuming equilibrium between macro and microstructural water, the chemical potential of the sample will increase rapidly. There will be a fast increment in the microstructural void ratio and an oversized swelling. If the sample is longer (Fig. 8 c), the modelled hydration is slower. When the kinetics of the macro-micro mass exchange is considered, that is, in M11 simulation, this high sensitivity to the sample height is not observed, and the results are consistent with the test data in all cases.

This behaviour is even more evident in Fig. 9, which presents the results obtained in the second example of the application of the model, which is an analysis of a test performed by Dvinskikh et al. (2009). They used nuclear magnetic resonance imaging to characterise the spatial distribution of the clay particles during the swelling of a sample in deionised water. The sample was made of purified Na-ion exchanged MX-80

bentonite. The test scheme was similar to the one shown in Fig. 7. The sample had an initial height of 3 mm and a diameter of 8 mm. The initial gravimetric water content was 23% and the initial dry density was 1.8 g/cm^3 .

In the simulations of this test, the definition of intrinsic permeability used to obtain Figs. 8 a, b and c was changed. Instead, the formulation and mean parameters proposed by Liu (2010) for a MX-80 Na-bentonite in diluted solutions were used. However, because the analysis was focused more on comparing the results of the models than on fitting the experimental results, the rest of the parameters were assumed to be equal to those used in the simulation of the previous tests. Thus, lower quality fits were obtained. Even so, the inclusion of the f_2 mechanism makes it possible to satisfactorily reproduce the experimental data trend (M11 results in Fig. 9). However, this is not the most relevant aspect highlighted by the figure. Because the test has a longer duration (approximately 300 days) than the previous ones (2 to 7 days), it is able to show the difficulty in simulating the real kinetics of the process by using a model that assumes equilibrium between macro and microstructural water (M00 results, Fig. 9). Therefore, a new simulation, M01, of the test presented by Dvinskikh et al. (2009) was performed, disregarding the interaction mechanism represented by f_2 (which is the meaning of the “zero” in M01, see Figs. 2 and 6, and thus $f_s=f_1$). However, the existence of a kinetic mass exchange term, c_m , was taken into account using Eqs. (6), (7) and (11) (the “1” in M01). When introducing the non-equilibrium macro-micro water mass exchange, the kinetics of the process is no longer controlled only by the saturation of the microstructure. The “shape” of the model results is improved (see Fig. 9). However, M01 still does not accurately reflect the real behaviour. This question is analysed in Figs. 10, 11 and 12.

In Figs. 10 a and b, the path “A-B-C-D” followed in the spaces s - p and p - q (q being the von Mises stress) has been drawn for a point close to the top of the sample. Regardless of whether M11 or M01 is considered, practically the same paths are obtained. In these figures, “A” defines the initial conditions, “B” indicates the macrostructural saturation, “C” identifies the start of the macrostructural yielding and “D” indicates the final failure situation (virtually zero stress state). When failure occurs (complete soil softening), it entails complete soil destructuration, which may be interpreted as the method by which an elastoplastic model incorporates the transformation of bentonite soil into a sol. To avoid numerical problems, the softening process was blocked when the mean stress was close to zero ($p = 0.002$ kPa was the lower bound).

The path “A-B-C-D” is also depicted in Fig. 11 a, where the evolution of the computed values of p_O , p_O^* and p_R in the top of the sample is represented. The paths followed by p_O in both models are almost the same (they overlap in Fig. 11 a). The same occurs to p_O^* , which, from point “B” (full saturation), follows the same path as p_O . The yield surface is reached (point “C”) before reaching e_{mFS} (point “E” in Figs. 11 b and 12). Therefore, the paths followed by p_R when using M11 and M01 coincide almost perfectly. The only difference is that the plastic macrostructural strains induced by f_2 cause the softening process to progress at a faster rate when the f_2 mechanism is introduced (M11) than when it is ignored (M01) (Fig. 11 b), thereby reaching point “D” earlier. However, this small difference in the stress path is linked to an important difference in the strain behaviour.

Fig. 12 illustrates the computed evolution of the microstructural and macrostructural void ratios at the top of the sample. Because the same formulation was used to define e_m , Eq. (2), the path followed by e_m is almost identical in both cases (Fig. 12 a). Nevertheless, the same is not true for the macrostructural void ratio. When “C” is

reached, the bentonite is already saturated (point “B”). Consequently, there is no evidence of macrostructural strains associated with changes in suction (see Fig. 12 b). Moreover, because the yield surface has already been reached, f_1 will be equal to 0. Thus, if only the interaction function f_1 is used (M01), from the yield point, all the deformations will be associated with the macrostructural dilatance. As can be observed in Fig. 12 b, these deformations are smaller than those caused by the microstructure, and they are not strong enough to reproduce the swelling of the sample (Fig. 9). Hence, the dilatant strains introduced by the macrostructural model over the course of the softening stage “C-D” are not able to reproduce the deformations caused by the destructuring of the macrostructure in an FS process throughout this stage. These deformations can be reproduced if the plastic mechanism defined by the interaction function f_2 is introduced. In this case, as observed in Fig. 12 b, the macrostructural strains obtained with M11 are much larger, and the model produces higher quality results when simulating the experimental free swelling processes.

5. CONCLUSIONS

The application of an elastoplastic model for the simulation of free swelling (FS) processes in bentonite is reviewed. The inclusion of the kinetics of the water mass exchange process between the macrostructure and the microstructure in the model is advisable. This addition, see Eq. (7), makes it is possible to obtain simulation results that have an evolution that is not only controlled by the advancing macrostructural saturation front.

However, while the inclusion of the water mass exchange kinetics improves the ability of the model to describe the FS processes, the results are also improved by taking into account the strains associated with the macrostructure destructuring when the

aggregates swell under FS conditions. This phenomenon is proposed as a new active plastic mechanism introduced by a new term, which increases the value of the interaction function defining the plastic macrostructural strains induced by microstructural effects. This term, which has been called " f_2 ", see Eq. (9), is defined as a function of the microstructural void ratio, e_m (see Fig. 6 a). Hence, the model includes the experimental observation that the macrostructural destructuring becomes more intense when the microstructural void ratio is higher.

The formulation should be improved as experience is gained in its application to the simulation of the FS processes. For instance, the shape of f_2 will need improvement. It is also likely that even the functional dependence between f_2 and e_m will have to be modified. Another aspect that should be addressed is how to relate the macrostructure destructuration to a potential soil particle release when the bentonite becomes a sol (soil structure failure). Moreover, it should be stressed that the application of the model to calcium dominated bentonite, or sodium dominated bentonite in saline solutions, should be analysed with more detail.

However, even with these issues, the results (Figs. 8 and 9) are encouraging. They show that the inclusion of the new plastic mechanism enhances the capacity of elastoplastic models to reproduce the FS processes. Therefore, the introduction of f_2 contributes to the unified characterisation of the saturated and unsaturated bentonite-free swelling using elastoplastic models.

APPENDIX A. LIST OF SYMBOLS AND UNITS

C Constitutive parameter synthesising the way in which the water mass transfer term changes as π^* approaches π_B . Units: $(\text{kPa}\cdot\text{s})^{-1}$

- c_m Water mass exchange between aggregates and macrostructural water. Units: $\text{kg} \cdot (\text{m}^3 \cdot \text{s})^{-1}$
- de_m Microstructural void ratio increment. Dimensionless
- $d\varepsilon_{mV}$ Microstructural volumetric strain increment. Dimensionless
- $d\varepsilon_{MV}^e$ Macrostructural elastic volumetric strain increment. Dimensionless. See Appendix B
- $d\varepsilon_{MV, m}^p$ Macrostructural plastic volumetric strain increment induced by microstructural effects. Dimensionless
- $d\varepsilon_{MV, m1}^p$ Macrostructural plastic volumetric strain increment caused by macrostructural packing. Dimensionless
- $d\varepsilon_{MV, m2}^p$ Plastic volumetric strain increment induced by aggregate destructuring in the macrostructure. Dimensionless
- e Total void ratio. Dimensionless
- e_m Microstructural void ratio. Dimensionless
- e_{mFS} Value of e_m at which the destructuring of the macro is assumed to begin. Dimensionless
- e_{mO} Microstructural void ratio when the swelling pressure is equal to 0. Dimensionless
- e_{mR} Microstructural void ratio corresponding to the largest destructuring rate. Parameter of the linear model of f_2 . Dimensionless
- F Yield function. Units: kPa^2 . See Appendix B
- f_c BExM interaction function in microstructural compression. Dimensionless
- f_s BExM interaction function in microstructural swelling. Dimensionless
- f_l Interaction function in microstructural swelling (denoted as f_s in BExM). Dimensionless

f_{1M}	Maximum value of f_1 . Dimensionless
f_2	New additional interaction function in microstructural swelling. Dimensionless
f_{2M}	Maximum value of f_2 . Dimensionless
g	Gravity constant. Units: $m \cdot s^{-2}$
H	Constitutive parameter defining the transfer coefficient at the end of the mass exchange process. Dimensionless.
k	Parameter defining the increase in cohesion with suction. Dimensionless. See Appendix B
k_m	Microstructural deformability parameter. Dimensionless
k_M	Isotropic intrinsic permeability of the macrostructural liquid. Units: m^2
K_p	“Stress bulk modulus”. Units: kPa. See Appendix B
K_s	“Suction bulk modulus”. Units: kPa. See Appendix B
m, n	Van Genuchten’s retention curve parameters. Dimensionless
m_m	Mass of microstructural water per unit volume. Units: $kg \cdot m^{-3}$
m_M	Mass of macrostructural water per unit volume. Units: $kg \cdot m^{-3}$
M	Slope of the critical state line. Dimensionless. See Appendix B
P_{ATM}	Atmospheric pressure. Units: kPa
P_G	Gas pressure. Units: kPa
P_L	Macrostructural liquid pressure. Units: kPa.
p	Net mean stress. Units: kPa
p_C	Reference stress. Units: kPa. See Appendix B
p_O	Net mean yield stress at the current suction. Units: kPa
p_O^*	Saturated pre-consolidation stress. Units: kPa.
p_R	Reference stress defined in Fig. 4. Units: kPa
p_{REF}	Material parameter used to define κ_s . Units: kPa. See Appendix B

p_{TOT}	Mean stress. Units: kPa
p_S	Increase in cohesion with suction. Units: kPa. See Appendix B
q	Von Mises stress. Units: kPa
q_M	Macrostructural water seepage. Units: $m \cdot s^{-1}$
r	Material parameter used to define the macrostructural soil compressibility. Dimensionless. See Appendix B
s	Macrostructural water matric suction. Units: kPa.
Sr_M	Macrostructural degree of saturation. Dimensionless
$Sr_{M,0}$	Macropore residual degree of saturation. Dimensionless
T	Absolute temperature. Units: K
v_m	Microstructural water flow vector. Units: $m \cdot s^{-1}$
v_M	Macrostructural water flow vector. Units: $m \cdot s^{-1}$
v_S	Soil skeleton's deformation rate. Units: $m \cdot s^{-1}$
W	Water molar mass. Units: $kg \cdot mol^{-1}$
z	Vertical coordinate. Units: m
α	Van Genuchten's retention curve parameter. Units: kPa^{-1}
α_{Sp}	Material parameter used to define κ_s . Dimensionless. See Appendix B
α_{SS}	Material parameter used to define κ_s . Dimensionless. See Appendix B
α_i	Material parameter used to define $\kappa(s)$. Units: kPa^{-1} . See Appendix B
β	Material parameter used to define the macrostructural soil compressibility. Units: kPa^{-1} . See Appendix B
$\kappa(s)$	Elastic stiffness for changes in the net mean stress evaluated at macrostructural suction s . Dimensionless. See Appendix B
κ_{i0}	Material parameter used to define $\kappa(s)$. Dimensionless. See Appendix B

- $\kappa_S(s, p)$ Elastic stiffness for changes in suction evaluated at macrostructural suction s and net mean stress p . Dimensionless. See Appendix B
- κ_{S_0} Material parameter used to define κ_S . Dimensionless. See Appendix B
- κ_M Macrostructural relative permeability. Dimensionless
- $\lambda(s)$ Slope of the virgin compression curve at macrostructural suction s .
Dimensionless
- $\lambda(0)$ Slope of the virgin compression curve for saturated conditions. Dimensionless
- $\mu_{VO}(T)$ Reference chemical potential of water vapour over a flat surface of pure water at absolute temperature T . Units: $\text{kJ} \cdot (\text{mol})^{-1}$
- μ_W Dynamic viscosity of water. Units: $\text{Pa} \cdot \text{s}$
- μ_m Chemical potential of microstructural water inside the aggregates. Units: $\text{kJ} \cdot (\text{mol})^{-1}$
- μ_M Chemical potential of macrostructural water. Units: $\text{kJ} \cdot (\text{mol})^{-1}$
- π Bentonite swelling pressure. Units: kPa
- π_B Boundary swelling pressure. Units: kPa
- ρ_W Water density. Units: $\text{kg} \cdot \text{m}^{-3}$
- $\partial/\partial t$ Time derivative operator. Units: s^{-1}
- ∇ Gradient differential operator. Units: m^{-1}

APPENDIX B. SUMMARY OF THE CONSTITUTIVE EQUATIONS

The strain behaviour of sodium bentonite is characterized by the evolution of both the microscopic volumetric strain and the macroscopic strain. As described in section 2, the definition of π_B in Eq. (6), the state function in Eq. (2) defining e_m and the kinetic mass exchange model of c_m in Eq. (7) define the constitutive background to describe the

microscopic volumetric strain. An associated plasticity approach at constant suction, based on the BBM, was adopted to compute the macroscopic strain. As indicated in Section 4, the formulation used in this paper is based on that presented by Alonso et al. (2011). Navarro et al. (2014) presented the details of its computational implementation. However, a brief review of the equations introduced may still be of interest. In this work, as it is conventional practice whenever BBM is applied, the yield function F was defined as:

$$[B.1] \quad F = q^2 - M^2(p + p_s)(p_o - p) = 0$$

where q is von Mises stress, M is the slope of the critical state line (the value in Table 1 was used in the simulations) and p is the net mean stress. The increase in cohesion with suction p_s was computed as:

$$[B.2] \quad p_s = k \cdot s$$

k being a constitutive parameter (see Table 1). The net mean yield stress at the current suction p_o was calculated with the expression:

$$[B.3] \quad p_o = p_c \left(\frac{p_o^*}{p_c} \right)^{\frac{\lambda(0) - \kappa(s)}{\lambda(s) - \kappa(s)}}$$

where p_c is a reference stress (see Table 1), and the evolution of the saturated pre-consolidation stress p_o^* is given by the hardening law defined in Eq. (8). The slope of the virgin compression curve at macrostructural suction s $\lambda(s)$ (Appendix A) was defined by the law:

$$[B.4] \quad \lambda(s) = \lambda(0) [r + (1 - r) \exp(-\beta \cdot s)]$$

where $\lambda(0)$, r and β are material parameters (Table 1). To define the elastic stiffness for changes in suction, κ_s , the expression by Hoffman et al. (2007) was adopted:

$$[B.5] \quad \kappa(s) = \kappa_{i0} \cdot (1 + \alpha_1 \cdot s)$$

κ_{i0} and α_i being material parameters (see Table 1).

The macrostructural elastic volumetric strain $d\varepsilon_{MV}^e$ was computed as:

$$[B.6] \quad d\varepsilon_{MV}^e = \frac{dp}{K_p} + \frac{ds}{K_s}$$

where the “stress” bulk modulus K_p was computed as:

$$[B.7] \quad K_p = \frac{p(1+e)}{\kappa(s)}$$

and the “suction” bulk modulus K_s was obtained with the expression:

$$[B.8] \quad K_s = \frac{(s + P_{ATM})(1+e)}{\kappa_s(s, p)}$$

$\kappa_s(s, p)$ is the elastic stiffness for changes in suction evaluated at macrostructural suction s and net mean stress p are computed with the law:

$$[B.9] \quad \kappa_s(s, p) = \kappa_{s0} \cdot \left(1 + \alpha_{sp} \cdot \ln\left(\frac{p}{p_{REF}}\right) \right) \cdot \exp(\alpha_{ss} \cdot s)$$

where κ_{s0} , α_{sp} , p_{REF} and α_{ss} are material parameters. The values used in the simulations are indicated in Table 1. The value adopted to define Poisson’s ratio ν is also indicated in Table 1, which, together with K_p and K_s , define the elastic behaviour of the bentonite.

APPENDIX C. AN INSPECTION ON SENSITIVITY ANALYSIS

Although an in-deep analysis of the sensitivity of the model to the new parameters is beyond the scope of this work, it is of interest to illustrate this sensitivity with an inspection exercise. The change in the total swelling of the sample has been analysed for the maximum and minimum values of the parameters in Table C.1. As can be observed, the maximum value “P_MAX” (“P” being e_{m0} , k_m , C , H , e_{mFS} or f_{2M}) has been taken approximately as twice its average value “P_Average”. P_Average corresponds to the value used to obtain Figs. 8 – 12. The minimum value “P_min” is approximately

half of the average value. Only one of the parameters is modified in each simulation. This way, for instance, the curve emO_min in Fig. C.1 a uses $e_{m0}=40$, while keeping the remaining parameters at their average values. The parameters considered for the sensitivity analysis are those defining the new laws introduced in this work: e_m , Eq. (2), c_m , Eq. (7), and f_2 , Fig. 6 b.

Figures C.1 represent the results obtained when simulating test T3 (Fig. 8 c) from B+Tech (sample initially 40 mm high). This figure shows that the microstructure deformability k_m plays the most important role in characterising the free swelling processes of sodium dominated bentonites.

For better illustration, the previous analysis was repeated with a smaller deviation of P_{MAX} ($=1.1 \cdot P_{Average}$) and P_{min} ($=0.9 \cdot P_{Average}$) from the average values. The solutions obtained were compared to the one obtained with the average values (“Average curve” in Fig. C.1). For that purpose, an error function equal to the root mean square deviation was used. This way, for P_{MAX} , the following was computed:

$$[C.1] \quad error_{P_{MAX}} = \sqrt{\frac{\sum_{i=1}^N (\delta_{AV,i} - \delta_{P_{MAX},i})^2}{N}}$$

where $\delta_{AV,i}$ is the total swelling value computed at time i when the average set of parameters is used, and $\delta_{P_{MAX},i}$ is the swelling computed at the same time when “P” parameter takes P_{MAX} value while the rest of parameters keep their average value. In the analysis, $N=12$ computational time steps were taken. The sensitivity of the model with respect to each parameter in the range considered, $sensitivity_P$, was estimated with:

$$[C.2] \quad sensitivity_P = MAX(error_{P_{MAX}}, error_{P_{min}})$$

The obtained sensitivity values have been represented in Fig. C.2. It confirms that, even in a local study in the neighbourhood of the average values considered, k_m is the parameter for which the model shows the greatest sensitivity.

ACCEPTED MANUSCRIPT

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FIGURES

Figure 1. (a) Schematic particle arrangement in a highly compacted Na-bentonite: 1, intra-aggregate space; 2, interaggregate space; and 3, aggregate. (b) State after swelling and saturation. Adapted from Pusch (1987).

Figure 2. Interaction functions between micro and macroporosity deformations. Adapted from Alonso (1998).

Figure 3. (a) Representation of the “loading- collapse” (LC) yield surface in the $p-s$ space. (b) Yield surface in the $p-q$ space and definition of the reference pressure p_R ; “CSL” defines the critical state line.

Figure 4. Variation of the aggregates void ratio in the tests carried out by Montes-H et al. (2003). Numbers in the legend correspond to the relative humidity values. “T” indicates experimental data and “M” indicates the model results.

Figure 5. Variation of the macroscopic (e_M) and microscopic (e_m) void ratio in several processes of free swelling.

Figure 6. (a) Function f_2 . (b) Function f_1 .

Figure 7. General configuration of the tests carried out by B+TECH and Dvinskikh et al. (2009).

Figure 8. Experimental and model results of the tests carried out by B+TECH. (a) Test 1, (b) Test 2, (c) Test 3.

Figure 9. Experimental and model results of the test carried out by Dvinskikh et al. (2009).

Figure 10. Computed paths for a point close to the top of the sample in the tests carried out by Dvinskikh et al. (2009). (a) s - p path, (b) p - q path.

Figure 11. (a) Evolution of the computed values of p_O , p_O^* and p_R in the top of the sample in the tests carried out by Dvinskikh et al. (2009). (b) Detail of the p_R paths.

Figure 12. Computed evolution of (a) e_m and (b) e_M at the top of the sample in the tests carried out by Dvinskikh et al. (2009).

Figure C.1. Model results when the parameters P_MAX and P_min in table C.1 are used.

Figure C.2. Sensitivities obtained according to Eq. (C.2).

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Figure C1

Figure C2

Table 1. Mechanical parameters used in the simulations shown in Figs. 8 to 12. The notation by Alonso et al. (2011) is adopted.

Parameter	Value
k	0.1
κ_{io}	0.05
α_i (1/kPa)	$-3.0 \cdot 10^{-6}$
κ_{So}	0.25
α_{Sp}	0
α_{SS} (1/kPa)	0
p_{REF} (kPa)	10
ν	0.35
p_C (kPa)	10
$\lambda(0)$	0.15
r	0.8
β (1/kPa)	$2.0 \cdot 10^{-5}$
p_O^* (kPa)	10400
M	1.07

Table C.1. Minimum, P_min, average, P_Average, and maximum, P_MAX, values for the “P” parameters considered in the sensitivity exercise (Fig. C.1).

Parameter “P”	Units	Average Value	minimum value “P_min”	MAXIMUM value “P_MAX”
e_{mo}	Dimensionless	21	10	40
k_m	Dimensionless	6.6	3	12
C	(kPa·s) ⁻¹	0.4	0.2	0.8
H	Dimensionless	$1.5 \cdot 10^{-8}$	$0.5 \cdot 10^{-8}$	$3.0 \cdot 10^{-8}$
e_{mFS}	Dimensionless	2.7	1	6
f_{2M}	Dimensionless	1	0.5	2

HIGHLIGHTS

- We propose a new elastoplastic mechanism to model bentonite free swelling.
- Plastic strains induced by microstructural effects are activated by free swelling.
- A new approach to the calculation of the microstructural void ratio is introduced.
- It is advisable to use a kinetic micro-macrostructural water exchange term.
- The proposed formulation yields satisfactory results for the analysed tests.