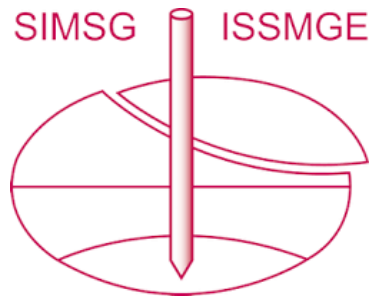


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New insights for modeling the mechanical behavior of clays under different environmental conditions

Nouvelles perspectives pour la modélisation du comportement mécanique des argiles dans différentes conditions environnementales

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ABSTRACT: The growing demand for environmentally-related geotechnical applications where clays are involved (e.g., nuclear waste storage repositories and geosynthetic liners) results in the need to formulate advanced geomechanical approaches. When subjected to variations in the degree of saturation and (or) chemical composition of the pore water, clays (especially if active) can show significant volume and shear strength changes. The present paper shows how accounting for the chemical interaction between clay particles and water implies reformulating the effective stress definition. Considering existing experimental data, this contribution provides some interpretation advantages gained when adopting the proposed concept. Prominent among these implications is the uniqueness of the failure envelope regardless of the chemical composition of the pore water, together with the ensured transition between unsaturated and saturated states. As a result of these implications, essential advantages for the stress-strain constitutive modeling of these geomaterials in challenging application contexts are expected.

RÉSUMÉ : La demande croissante d'applications géotechniques liées à l'environnement impliquant des argiles (p. ex., les dépôts de déchets nucléaires et les revêtements géosynthétiques) entraîne la nécessité de formuler des approches géomécaniques avancées. Lorsqu'elles sont soumises à des variations du degré de saturation et (ou) de la composition chimique de l'eau interstitielle, les argiles (surtout si elles sont actives) peuvent présenter d'importants changements de volume et de résistance au cisaillement. Le présent article montre comment la prise en compte de l'interaction chimique entre les particules d'argile et l'eau implique de reformuler le concept de contrainte effective. En considérant les données expérimentales existantes, certains avantages d'interprétation obtenus en adoptant le concept proposé sont fournis dans cette contribution. L'une des principales implications est l'unicité de l'enveloppe de rupture, quelle que soit la composition chimique de l'eau interstitielle, ainsi que la transition assurée entre les états non saturés et saturés. En conséquence de ces implications, des avantages essentiels pour la modélisation constitutive contrainte-déformation de ces géomatériaux dans des contextes d'applications complexes sont attendus.

KEYWORDS: clay, effective stress, matric suction, chemical composition, pore water

1 INTRODUCTION

Environmental conditions may vary the mechanical response of clays (especially if active) very significantly (e.g., Di Maio, 1996; Manca et al., 2016). Consequently, modeling the mechanical behavior of clays in advanced engineering applications requires to devote particular attention to the definition of (i) relevant stress variables, including those representatives of the hydraulic and chemical states; (ii) appropriate strain variables; (iii) proper constitutive stress-strain relationships, accounting also for chemo-hydro-mechanical couplings.

In the present paper, first, the effects (on the structure of the water and the types of ions) of the chemical interaction between clay particles and water are identified. Then, two of the main existing formulations of effective stress for saturated and unsaturated soils are recalled. Subsequently, an appropriate effective stress concept for clays is described. It includes the possibility of accounting for the effects on the mechanical stress-strain response of capillarity, chemical interaction between solid particles and water, and bulk water chemical composition. It is shown that in the most general case - in addition to the effective stress definition and the definition of the corresponding stress-strain constitutive model - there is a need for the matric suction stress variable, a chemical stress variable, and the relationships describing the evolution of both the latter variables at varying degree of saturation. Several simplifications are gained when adopting the proposed effective stress concept. Among them, by referring to residual or critical states, it is revealed the uniqueness of the shear strength parameters irrespective of the chemical

composition of the pore water and the independence of the saturation state. These interpretation advantages pose the basis for a convenient stress-strain constitutive modeling approach.

2 DEFINITIONS OF WATER AND IONS IN CLAYS

Clay particles are characterized by multiple basic units consisting of two or three different sheets (Beek et al., 1978; Mitchell and Soga, 2005). The external surfaces of clay particles exhibit a negative charge deficit due to the (partial or total) dissociation of cations in the presence of water (Beek et al., 1978) (Figure 1). Although replaceable by other cations, these dissociated cations cannot leave the clay and can extend to distances from particle surfaces of even more than 1 nm (Tournassat et al., 2009), up to 50 nm according to (Terzaghi et al., 1996). They are named *non-movable ions*; all other cations or anions in water that are not the result of interactions with solid surfaces are *movable ions* (Tuttolomondo et al., 2021) (Figure 1). The negatively charged surfaces of the clay particles affect the structural characteristics of the water in the vicinity of the clay particles. Laboratory evidence and numerical simulations reveal an ordered water structure near clay mineral surfaces (Cheng et al., 2001; Park and Sposito, 2002; Mante et al., 2014; Zhao et al., 2015; Chen et al., 2018). Studies on water density, viscosity, elastic properties, and diffusion coefficient show that water properties vary only within 1 nm from the clay mineral surfaces (Tournassat et al., 2009; Mante et al., 2014; Adapa and Malani, 2018). Due to its structural peculiarities, water within this distance range can be defined as *solid water* (Tuttolomondo et al., 2021); water beyond 1 nm can be named accordingly as *liquid water* (Figure 1). Since liquid

water can be found starting at 1 nm from the particle surfaces, it is evident that it can include both movable and non-movable ions (Figure 1). *Diffuse layer* and *bulk water* are defined as those water layers that include only non-movable ions and movable ions, respectively (Figure 1). The deficit in the negative charge of clay particles and the consequent need to distinguish between solid water, liquid water, non-movable ions, and movable ions are the more relevant, the more active the clay minerals are (see Skempton, 1953 for the definition of activity).

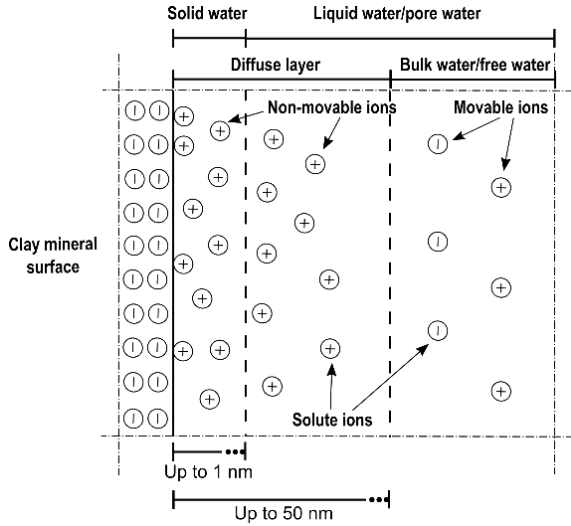


Figure 1. Definition of water and ions in clays (redrafted from (Tuttolomondo et al., 2021)).

3 EFFECTIVE STRESS CONCEPT FOR CLAYS UNDER DIFFERENT ENVIRONMENTAL CONDITIONS

An appropriate effective stress framework can enable converting a real medium characterized by n mixtures (e.g., solid mixture and fluid mixtures) and subject to m actions (e.g., total stress changes and environmental actions) into a continuous medium, mechanically equivalent (i.e., with the same solid skeleton deformation and strength change) subject to a change in effective stress. The framework is dependent on both the geomaterial and the type of actions to be considered for the specific geomechanical problem of interest.

In the following, the effective stress variable and, eventually, additional stress variables will be introduced for different soils and saturation states. When needed, the stress-strain relationships for the additional stress variables will also be discussed. In contrast, the stress-strain constitutive relation for the solid skeleton is not the object of the present paper.

For soils characterized by two different mixtures - the solid mixture and liquid water - (saturated soils), the Terzaghi's effective stress reads (Equation 1):

$$\sigma'_{ij} = \sigma_{ij} - u_w \delta_{ij} \quad (1)$$

where σ_{ij} is the total stress tensor, and $u_w \delta_{ij}$ is the measured (or externally imposed) water pressure tensor.

For soils characterized by three different mixtures (unsaturated soils) - the solid mixture, liquid water, and a gaseous solution - one of the possible formulations of the effective stress variable is as follows (Nuth and Laloui, 2008b) (Equation 2):

$$\sigma'_{ij} = \sigma_{net,ij} + S_r s_m \delta_{ij} \quad (2)$$

where $\sigma_{net,ij} = \sigma_{ij} - u_g \delta_{ij}$ is the net stress tensor (being u_g the gaseous pressure), $s_m \delta_{ij} = (u_g - u_w) \delta_{ij}$ is the matric suction stress tensor and S_r is the degree of saturation. For saturated states, Eq. 2 recovers Eq. 1. For Eq. 2 to be suitably solved, the dependence of s_m on S_r can be addressed by employing a proper soil water retention model.

In the case of soils whose solid mixture includes clay minerals, the definition of effective stress for saturated and unsaturated states can be reformulated to account for the chemical interaction between clay particles and water molecules explicitly (see Figure 1).

In particular, a thermodynamic approach coupled with geochemical principles with consideration of both non-movable and movable ions allowed formulating the generalized effective stress concept for saturated active clays (Tuttolomondo et al., 2021) and defining the corresponding effective stress variable as follows (Equation 3):

$$\sigma'_{ij} = \sigma_{ij} - (u_w + s_{s,e}) \delta_{ij} \quad (3)$$

where $s_{s,e} \delta_{ij}$ is the effective solute suction stress tensor. The effective solute suction is the difference between the solute suction of the pore water and the solute suction of the bulk water. The former depends on the presence of both non-movable and movable ions; the latter relies on the presence of movable ions only. This chemical variable can be determined using different methods (Tuttolomondo et al., 2021). The variable $s_{s,e}$ will assume greater values the more active the clay is. For non-active clays, or, more generally, for geomaterials where the interactions between solid particles and water are not relevant, Eq. 3 recovers Eq. 1.

Applying a methodology similar to (Tuttolomondo et al., 2021) but referring to unsaturated states allows updating Eq. 2 for clays as follows (Equation 4):

$$\sigma'_{ij} = \sigma_{net,ij} + S_r (s_m - s_{s,e}) \delta_{ij} \quad (4)$$

For Eq. 4 to be suitably solved, the dependence of s_m and $s_{s,e}$ on S_r must be appropriately addressed. Figure 2 shows the analytically determined dependence of $s_{s,e}$ on S_r for Ponza bentonite (Di Maio, 1996) (cation exchange capacity equal to 74 cmol/kg; fraction of non-movable cations within the pore water equal to 0.15 (Tuttolomondo et al., 2021)), assuming a void ratio equal to 1, a solid unit weight equal to 2.67 Mg/m³ and a 0.2 M NaCl bulk water. In the same figure, a power function of the type $s_{s,e} = a S_r^b$ is proposed and will later be shown to be of interest (parameters adopted in Figure 2: $a=369$ kPa and $b=-1.27$). For saturated states, Eq. 4 recovers Eq. 3.

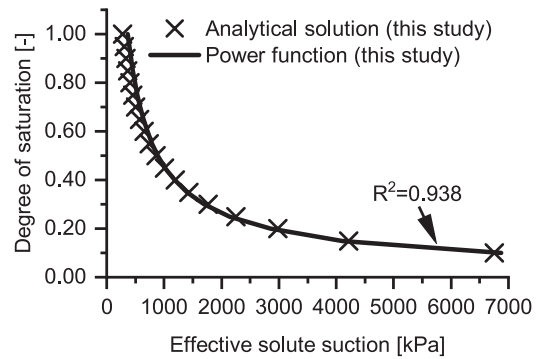


Figure 2. Dependence of $s_{s,e}$ on S_r for Ponza bentonite: analytical solution and power function.

4 SOME IMPLICATIONS FOR THE SHEAR STRENGTH ENVELOPE

In this section, some implications of adopting the developed effective stress concept for clays will be described. In particular, in the present paper, the attention is limited to those concerning the shear strength data for specimens tested under saturated and unsaturated conditions. Further studies and details for saturated states are provided in Tuttolomondo et al. (2021).

4.1 Saturated conditions

Di Maio reported the results of residual shear strength of Ponza bentonite (clay fraction of 80%, kaolinite and Na-smectite fractions of approximately 20% and 80%, respectively; plasticity index with distilled water equal to 320%, limit liquid with distilled water equal to 390%; Di Maio (1996) and Di Maio et al. (2004)). Specimens were prepared (at their liquid limits) and tested using different solutions (distilled water and NaCl solutions up to 6.1 M). Figures 3a and 3b show the shear strength data following Terzaghi's effective stress (Eq. 1) and the generalized effective stress for active clays (Eq. 3), respectively. In each figure (Fig. 3a and 3b), the Mohr-Coulomb failure envelope, the shear strength parameters, and the coefficients of determination are reported. The comparison reveals an improvement in favor of the generalized interpretation. Within the latter framework, for each experimental point, $s_{s,e}$ was calculated as the difference between the solute suction of the pore water and the solute suction of the bulk water. The former was determined using an analytical approach (cation exchange capacity equal to 74 cmol/kg; fraction of non-movable cations within the pore water equal to 0.15) (Tuttolomondo et al., 2021); the latter was determined through an empirical function proposed based on the data reported by Lang (1967) and Witteveen et al. (2013) (Tuttolomondo et al., 2021) (Fig. 3c). Fig. 3d provides the calculated $s_{s,e}$ values and shows how $s_{s,e}$ decreases as the concentration increases, being the 0-0.5 M the range where a salt concentration change can play a substantial role. The same most significant range of concentration was highlighted in the literature by referring, for instance, to liquid limit data (Di Maio and Scaringi, 2016), compression index (Di Maio et al., 2004), oedometer modulus (Witteveen et al., 2013), shear strength (Di Maio, 2004; Di Maio et al., 2016) and swelling behavior (Manca et al., 2016)

4.2 Unsaturated conditions

Rosone et al. (2016) reported the results of matric suction controlled triaxial shear tests performed on compacted scaly clay from Sicily (liquid limit in the range of 60-64%, plastic limit in the range of 20-26%). Specimens were isotropically consolidated at mean net stress of 50, 100, and 200 kPa and subject to matric suction in the range of 50-500 kPa. Figure 4a provides the shear strength data according to Eq. 2 and Eq. 4. The shear strength envelope referred to saturated data only, and the coefficients of determination of the shear strength envelope referred to both interpretations are also provided in the same figure. For both interpretations, the soil water retention data (Rosone et al., 2016) were modeled according to Nuth and Laloui (2008a) (Fig. 4b). Additionally, for the interpretation with Eq. 4, the Mohr-Coulomb shear strength parameters referred to the saturated data were considered valid; then, by knowing all the other variables (degree of saturation, matric suction, shear strength, and mean net stress), $s_{s,e}$ values were back predicted. The back predicted values against the corresponding degree of saturation are shown in Fig. 4b together with the interpretation according to the proposed power function (see Figure 2). The data for the mean net stress of 50 and 100 kPa were treated together due to their similarity. The adopted parameters are provided in Table 1.

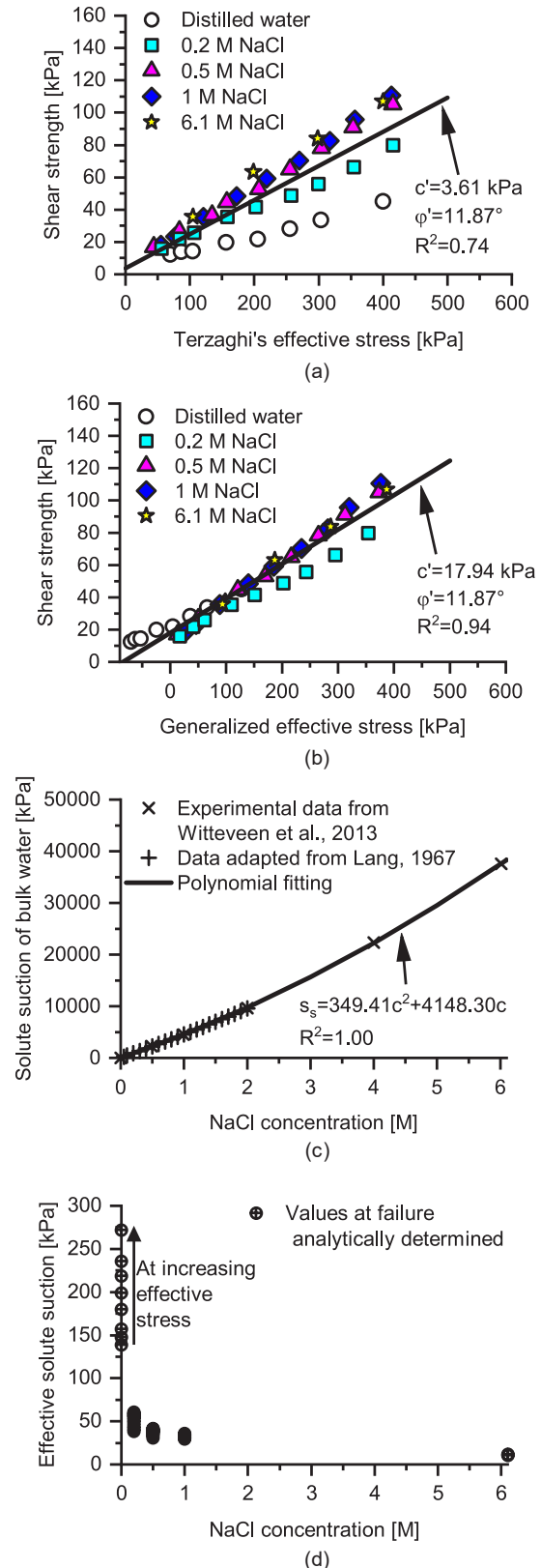
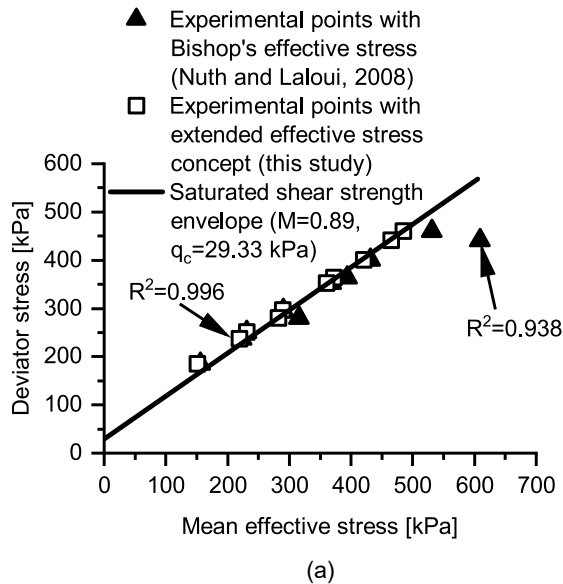


Figure 3. Shear strength data interpretation of Ponza bentonite: (a) Terzaghi's effective stress (redrafted from Tuttolomondo et al., 2021); (b) generalized effective stress for active clays (redrafted from Tuttolomondo et al., 2021); (c) polynomial fitting for solute suction of bulk water being c the NaCl concentration and s_s the solute suction of the bulk water (redrafted from Tuttolomondo et al., 2021); (d) effective solute suction at failure. Shear strength experimental data from Di Maio, 1996.

Figure 4a and 4b show, on the one hand, an improvement in the interpretation of shear strength data when using Eq. 4 (together with a properly defined $s_{s,e} - S_r$ and $s_m - S_r$ relationships) instead of Eq. 2 (together with the same properly defined $s_m - S_r$ relationship); on the other hand, the suitability of the power function (see Fig. 2) in capturing the variation of $s_{s,e}$ with S_r is also revealed.



- Back-predicted $s_{s,e}$ at $p_{net}=50$ kPa and 100 kPa
- Back-predicted $s_{s,e}$ at $p_{net}=200$ kPa
- ⊕ Soil water retention data at $p_{net}=50$ kPa and 100 kPa
- ⊞ Soil water retention data at $p_{net}=200$ kPa
- SWRC model (Nuth and Laloui, 2008)
- ESSC model (this study)

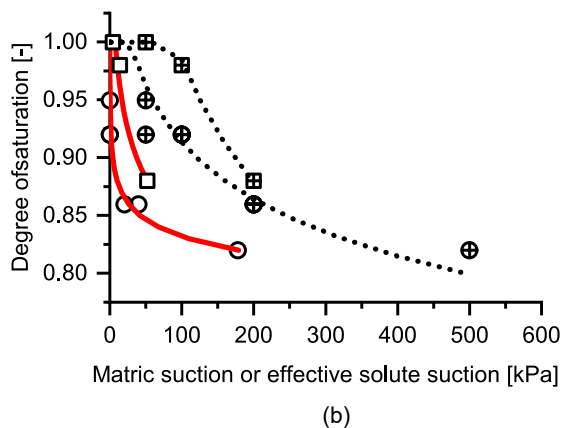


Figure 4. Shear strength data interpretation of scaly clay: (a) failure envelope according to two approaches; (b) Data and models of soil water retention and effective solute suction curves. Experimental data from Rosone et al., 2016 (p_{net} : mean net stress; SWRC: soil water retention curve; ESSC: effective solute suction curve).

Table 1. Parameters of the soil water retention curve and effective solute suction curve for the shear strength interpretation of the scaly clay.

Soil Water Retention Curve model parameters	$p_{net}=50$ kPa and 100 kPa	$p_{net}=200$ kPa
s_e [kPa]	22	60
S_{res} [-]	0.2	0.2
K_H [kPa]	3.90E+07	3.40E+07
β_H [-]	-22	-8
s_{D0} [kPa]	35	100
Effective Solute Suction Curve model parameters	$p_{net}=50$ kPa and 100 kPa	$p_{net}=200$ kPa
a [kPa]	0.06	7.81
b [-]	-40.41	-14.88

5 CONCLUSIONS

This paper presented new insights for modeling the mechanical behavior of clays under different environmental conditions, accounting for the chemical interaction between clay particles and water. The modified effective stress variable for saturated and unsaturated states is provided. The framework introduces the chemical variable named effective solute suction and an appropriate relationship between the latter and the degree of saturation. Some of the verified implications of using the proposed approach for saturated and unsaturated soils are presented. In particular, by referring to an active clay, the residual shear strength parameters were found to be independent of the chemical composition of the pore water; by referring to a non-active clay, the independence of the critical shear strength parameters of the saturation state was verified. The variation of effective solute suction during the considered tests is consistent with the expectations. It captures the scientific evidence of a higher mechanical impact of a chemical composition change in a range of concentrations below 0.5 M.

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