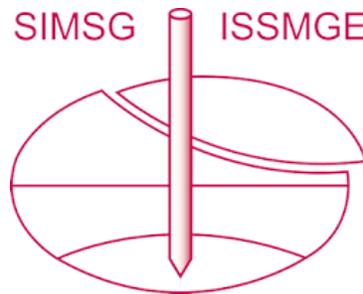


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An Effective Stress Framework for Clayey Geomaterials Including a Chemical-Dependent Variable

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Abstract. Experimental evidence shows that the chemical composition of the liquid inside the pores and its variation over time can have significant implications on the mechanical and hydraulic behavior of clayey geomaterials. The magnitude of these implications depends on the characteristics of the geomaterial and on the conditions to which it is subjected. This paper develops an effective stress model which includes a chemical-dependent variable. Some experimental results reinterpreted using the proposed effective stress and the generalized Bishop's effective stress are presented in order to show some potentialities of the proposed approach.

Keywords. Effective stress, chemistry, salt concentration.

1. Introduction

The chemical composition of the pore liquid within the geomaterials (commonly and simplistically identified as *pore water*) and its possible variability over time is a key topic for the geomechanical analysis in different applications. Among these, of particular importance are those in the field of slope stability (a decrease in shear strength as a result of pore liquid changes is considered one of the possible causes inducing the activation or reactivation of landslides [e.g. 1]), Geosynthetic Clay Liner (the chemical composition of the liquid in the subsoil may affect the hydraulic performances of GCLs [e.g. 2]), petroleum industry (the chemical design of the drilling fluids is fundamental to avoid instability phenomena in wellbores [e.g. 3]) and nuclear waste storage (the hydro-mechanical behavior of one or more barriers constituting the repository may change depending on the variation in the chemical composition of their pore-liquids [e.g. 4]).

Experimental evidence showing the influence of chemical composition on the mechanical behaviour of geomaterials is available in literature with reference to volumetric behaviour and shear strength. Experimental tests are usually conducted on reconstituted or undisturbed specimens and the main effects investigated concern the influence of a change in the concentration of the substances within the solution and/or a change in its pH [5-7]. With particular reference to concentration of salts, shear

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strength tests show that the shear strength increases as the concentration of salt grows [8-10]. Moreover, in the case of drained processes, shear strength will depend on the initial chemical composition of the solution within the geomaterial and on the way it will vary during the process [8]. With reference to volumetric results, a decrease in the compression index and the swelling index is noticeable as the concentration increases [11, 12]. In addition, an increase in salt concentration under constant total stress results in consolidation, whereas a decrease implies swelling [13-16]. The reversibility of these processes associated with an inverse chemical path is dependent on the mineralogy of the geomaterial and on the chemical characteristics of the involved liquid solutions compared to mineralogy [17, 18]. The magnitude of the main experimental observations briefly summarized here is particularly dependent on the presence of clay minerals and their characteristics. For example, as the bentonite content (i.e. mainly montmorillonite) in a mixture of kaolinite-bentonite increases, the differences in the plasticity index of the mixture, obtained with different salt concentrations, are more relevant [19].

Clay minerals are characterized by a crystalline structure. The electroneutrality of the dry mineral is guaranteed by the presence of cations on the external surfaces [20]. The mole number of cations on the outer surface depends on the negative charge the structure would have in the absence of them. An estimation of the quantity of these cations, referred not to the single particle, but to a representative elementary volume, is obtained thanks to the cation exchange capacity.

Examples of effective stress models including a variable dependent on chemistry are those developed at the scale of the particles (a review of those developed up to 1992 is provided in [21]). These models, while showing potentialities in the qualitative interpretation of observed phenomena, are of limited practical use due to the fact that the variables defined are relative to a scale much lower than that of the representative elementary volume. A more recent model, developed at the scale of the representative elementary volume, is that of Wei [22] in which work all the cations necessary for balancing the negative charge of the particles are implicitly considered part of the liquid solution.

The aim of this paper is to develop a definition of effective stress for clay geomaterials with a focus on taking into account clay minerals-water interactions. In particular, a fraction of cations, dissociated from the clay mineral's surfaces but part of the liquid water, is considered in the development procedure. Some implications of the proposed framework are verified by reinterpreting existing experimental results.

2. Development of the framework

The geomaterial is considered as a thermodynamic system characterized by two fluid solutions (a liquid solution and a gaseous solution) and a non-homogeneous mixture of solids which include clay particles. The solutions are, by definition [23], homogeneous mixtures within which it is possible to distinguish a substance present in greater quantities (conventionally called solvent) and substances present in smaller quantities (conventionally called solutes).

The solutions within the geomaterial consist of several chemical species and the quantity of each species inside each mixture can be expressed in terms of mole

numbers (n_i mole numbers of the species i which may vary from 1 to M_i , where M_i is the solubility of the species i).

According to the proposed framework, within the liquid solution, the solvent is water and the solutes are ions that can be categorized in two different classes:

- a) Ions related to the dissociation of salts; these ions are not necessary to ensure the electroneutrality of clay particles;
- b) Positive ions, dissociated from the clay mineral's surfaces, attracted by the particles but placed at a sufficient distance to be part of the water in its liquid state.

The hypothesis of homogeneity of the mixture, implicit in the definition of liquid solution, implies a homogeneous distribution of the solutes within the solvent. The positive ions which are necessary to guarantee the electroneutrality of clay particles but which are strongly attracted by them are considered part of the mixture of solid substances.

The water pressure within the geomaterial can be measured or imposed through different devices whose characteristics are also dependent on the pressure range of interest (positive or negative). Manometers are an example of devices for measuring the pressure at a point. The fluid present in the system of interest is balanced by a column of fluid of the same type or of a different type than the one in the system [24].

In the present study the water pressure at a point of the representative elementary volume is ideally measured thanks to a manometer connected to the geomaterial through a membrane permeable to both ions and solutes (if the pressure range is negative, this type of device is in the literature identified as a device for measuring the matric suction). The presence of solutes of type (b) within the liquid solution of the geomaterial implies that the piezometric surface identified by the device does not coincide necessarily with the piezometric surface of the liquid solution within the geomaterial. The device and the geomaterial are electrically neutral systems in thermodynamic equilibrium and, as a consequence, with equal total head.

The differential form of the Gibbs energy (G) for a generic mixture with pressure, temperature and composition variable during a transformation is called fundamental equation of chemical thermodynamics [25] and it is given by Eq. (1)

$$dG = \left(\frac{\partial G}{\partial T} \right)_{p, n_i} dT + \left(\frac{\partial G}{\partial p} \right)_{T, n_i} dp + \left(\frac{\partial G}{\partial n_1} \right)_{T, p, n_j} dn_1 + \left(\frac{\partial G}{\partial n_2} \right)_{T, p, n_j} dn_2 + \dots, \quad (1)$$

where T is the temperature and p is the pressure; the notation n_i is used when the partial derivative is calculated by considering that all the mole numbers of each constituent of the mixture are constant; the notation n_j is used when the partial derivative is calculated with respect to the mole numbers of one constituent when all the others are constant. For the partial derivatives of Eq. (1), the relations given in Eq. (2) are valid [25]:

$$\left(\frac{\partial G}{\partial T} \right)_{p, n_i} = -S \quad \left(\frac{\partial G}{\partial p} \right)_{T, n_i} = V \quad \left(\frac{\partial G}{\partial n_i} \right)_{T, p, n_j} = \mu_i \quad (2)$$

in which S is the entropy, V is the volume and μ_i is the partial molar Gibbs energy (or chemical potential), of the constituent i of the mixture.

By imposing the conditions of thermodynamic equilibrium in terms of chemical potential (Eq. (2)) at two different points at the same geodetic level (with respect to a chosen reference system) but one within the liquid solution of the geomaterial P_1 and one within the device P_2 , it is possible to determine the expression of water pressure within the geomaterial ($u_{w,P1(geom)}$) (Eq. (3)),

$$u_{w,P1(geom)} = u_{w,P2(man)} + \frac{RT}{\bar{V}} \ln \frac{x_{w,P2(man)}}{x_{w,P1(geom)}} \quad (3)$$

where $u_{w,P2(man)}$ is the water pressure measured by the manometer, $x_{w,P2(man)}$ is the molar fraction of water within the manometer, $x_{w,P1(geom)}$ is the molar fraction of water within the geomaterial, R is the universal gas constant and \bar{V} is the molar volume of water. The second term in the second member of Eq. (3) is called effective solute suction and it represents the chemical dependent variable within the proposed framework. By referring the quantity obtained in Eq. (3) to the entire representative elementary volume and by replacing it within the generalized Bishop's effective stress definition [26], the expression of effective stress given in Eq. (4) is obtained,

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

where $\sigma_{net,ij}$ is the net stress tensor defined as the difference between the total stress tensor σ_{ij} and the air pressure tensor $u_a\delta_{ij}$, S_r is the degree of saturation, s_m is the matric suction and $s_{o,e}$ is the effective solute suction.

3. Results

In this section, the reinterpretation of some experimental data available in literature is performed by using the effective stress definition given in Eq. (4). In addition, the interpretation of the same data according to the generalized Bishop's effective stress [26] is provided.

The experimental data referred to in this section are those obtained from the shear strength tests provided by [15]. The shear strength tests were carried out by the author using the classic direct shear apparatus. The geomaterial tested is a bentonite (clay fraction equal to 0.80, kaolinite fraction equal to 0.20 and Na-smectite fraction equal to 0.7-0.8). The author performed tests on reconstituted specimens using different solutions of different liquid compositions. In particular, the solutions used were distilled water and sodium chloride solutions with concentrations of 0.2 M, 0.5M, 1M and 6.1M. The samples were preliminarily reconstituted at a water content equal to their corresponding liquid limit, subjected to a stress of 400 kPa and unloaded at different stresses in order to perform direct shear tests at different stress level.

The direct shear tests data are in this work interpreted using the Mohr-Coulomb shear strength criterion with the corresponding effective stress of interest. The linear interpolation models, adopted for the whole stress range considered by the author [15], are in order to ensure that the coefficient of determination associated with them is greater than 0.90.

The results obtained, expressed in terms of shear strength angle and intercept cohesion for different values of sodium chloride concentrations, are shown in Figures. 1 and 2, respectively.

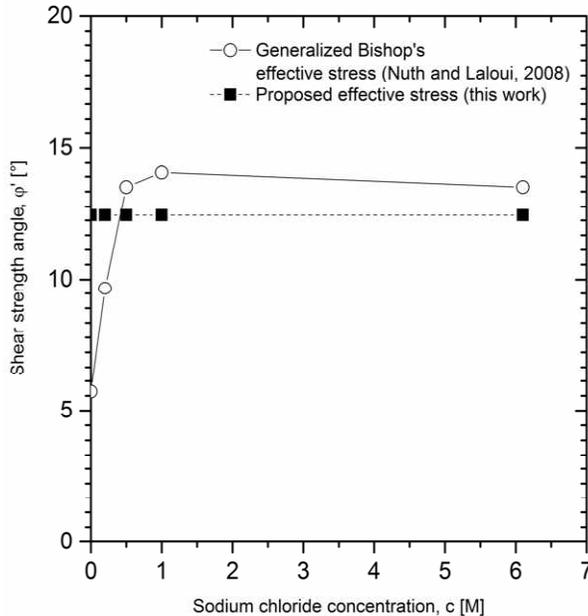


Figure 1. Shear strength angle of a bentonite for different values of salt concentration. Experimental data from [15].

In the case in which the results are interpreted by using the generalized Bishop's effective stress [26], it is possible to highlight a change in shear strength angle as the concentration varies. In particular, in the range of concentrations between 0 M NaCl (distilled water) and 1 M NaCl, the shear strength angle varies in the range between 6 ° and 15 °. As regards the variation of the cohesion intercepts with respect to the salt concentration, it is evidenced a not monotonic trend with values varying in the range between 5 kPa and 13 kPa.

If the results are interpreted by using the effective stress proposed in Eq. (4), instead, the linear interpolation model adopted with reference to the entire data set available allows to obtain a coefficient of determination greater than 0.90. For this reason, it is possible to observe from Figures. 1 and 2 that the value assumed by the shear strength angle and the intercept cohesion results to be independent on the concentration of the solution used to reconstitute and test the bentonite under examination.

4. Final remarks

According to the proposed approach, if there are no clay particles characterizing the solid fraction of the geomaterial of interest, the contribution of ions constituting category (b) of the solutes will be zero. Therefore, in this specific case, the

concentration of solute ions in the solution within the device for measuring the pressure and the concentration of solute ions in the liquid solution within the geomaterial will be the same. It follows that the second term in the second member of Eq. (3) will be zero and Eq. (4) will coincide with the generalized Bishop's effective stress [26].

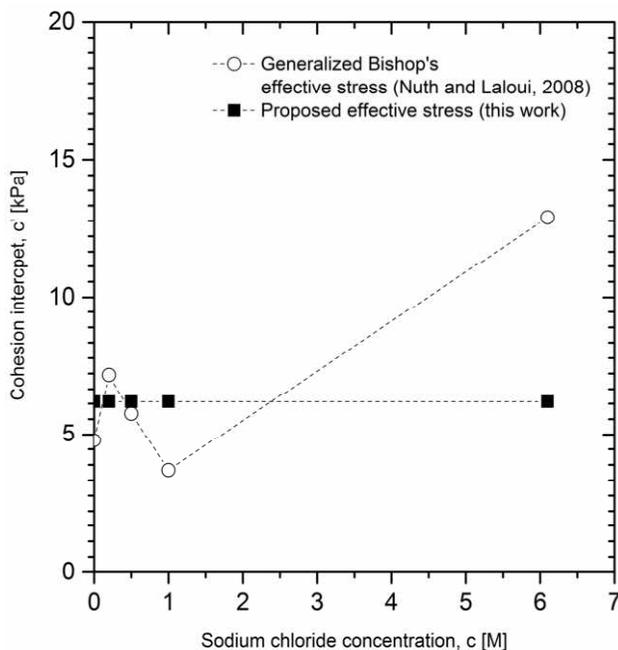


Figure 2. Intercept cohesion of a bentonite for different values of salt concentration. Experimental data from [15].

The dependence of the effective shear strength parameters on the concentration, which is obtained if the generalized effective stress is adopted (it coincides with the Terzaghi's one for a saturated medium), is reported in several works with reference to other geomaterials [8, 27].

The effective stress model developed in this work introduces a variable dependent on the chemical composition of the liquid solution (called effective solute suction). A comparison between the interpretation of some experimental data using the generalized Bishop's effective stress and the proposed definition has allowed highlighting the advantages in using the latter. Moreover, the proposed definition has the potential to be used within an elastic or elasto-plastic constitutive formulation.

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