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Unified characterization of hydro-mechanical properties of soil-bentonite mixtures exposed to pore-fluid salinity^{*#}

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1 Introduction

Knowledge on the impact of pore-fluid salinity, characterized by the changes of pH, ionic concentration, material composition, and other factors, on geotechnical engineering applications involving hydro-chemo-mechanical systems, is expanding. These systems range from conventional geotechnical applications (e.g. dams, hydraulic fills or cutoff walls) to geo-environmental facilities (e.g. deep geological disposal for radioactive waste, landfills) (D'Appolonia, 1980; Ismeik et al., 2013; Mishra et al., 2015). A favorable soil of bentonite was often adopted as an admixture in these projects owing to its high swelling and self-sealing capabilities (Ye et al., 2010; Zhang and Kröhn, 2019). Among a list of parameters, hydraulic conductivity (k) and undrained shear strength (S_u) are the most fundamental ones that are closely

related to the engineering performance of sealing materials (Dolar and Trauner, 2007; Deng et al., 2011; Chapuis, 2012). The characterization of these two parameters for soil-bentonite mixtures, while taking into account the influence of pore-fluid salinity, is therefore a matter of concern at both academic and practical levels.

Previous studies considering the effect of pore-fluid salinity often focused on pure clay (i.e. kaolinite, bentonite, and other types). A good example demonstrated by Zhu et al. (2013) showed that ionic-affected bentonite yields higher hydraulic conductivity (k). A similar trend was also found by Horpibulsuk et al. (2011), and the change of k for bentonite clay with different iron concentrations was interpreted using a generalized stress state (e/e_L). In doing so, the associated empirical correlation was given in a general form as


$$\frac{e}{e_L} = \alpha + \beta \log(k), \quad (1)$$

where e is the void ratio, e_L is the void ratio of bentonite clay at liquid limit, and the two constants are $\alpha=2.455$ and $\beta=0.229$. Moreover, Dolar and Trauner (2007) performed experiments on kaolinite clay with the addition of salt (NaCl). They found that, at the same water content, the undrained shear strength of specimens in deionized water was greater than that in saline solution; the authors further attributed the differences to the change of clay particle arrangements that resulted from interparticle forces. In contrast, Wahid et al. (2011) found that the residual strength of kaolinite was not sensitive to changes in iron concentration in the saline solution. Characterizing the hydro-mechanical properties of sand- (or silt-) clay mixtures and the associated particle interactions having various sources of complexity,

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compared to those for pure clay, is a significant scientific challenge (Mollins et al., 1996; Sivapullaiah et al., 2000; Yang and Liu, 2016; Deng et al., 2017; Liu et al., 2020b). Some researchers have studied the influence of ion concentration on saturated loess, which is primarily composed of silt and clay (Liu et al., 2019). It was found that the hydraulic conductivity of loess increases with the add-in salts in the solution (Xu et al., 2020). As far as soil-bentonite mixtures are of concern, the current literature is still lacking in a relevant method to evaluate k and S_u that accounts for the effect of pore-fluid salinity. Considerable interest remains in exploring whether the characterization of these two parameters can be achieved in varied pore-fluid conditions for the soil-bentonite mixtures.

In view of the above, this paper presents an experimental investigation of soil-bentonite mixtures, with the aim to provide an insight into changes of k and S_u by considering the effect of pore-fluid salinity. Discussions are made by comparing test results for specimens in different types of saline solution along with detailed interpretations at the micro-scale. More importantly, new correlations are established, enabling the unified characterization of k and S_u for soil-bentonite mixtures in varied conditions of pore-fluid salinity. Test results are presented in the section below. Readers are referred to Data S1 for the test procedures.

2 Results and discussion

2.1 Hydraulic conductivity (k) and undrained shear strength (S_u)

The hydraulic conductivity (k) and the undrained shear strength (S_u) of mixtures with different bentonite contents (B_c) were compared at a similar void ratio, as shown in Fig. 1. The number in the parentheses hereinafter denotes saline solution concentration (i.e. salt content by mass). It is readily observed in the figure that k decreases and S_u increases with the increase of bentonite content. More importantly, these changes are likely controlled by the types of pore fluid in the saturated specimens. Noticeably lower k values were found for specimens saturated with deionized water than that with saline solution. At $B_c=20\%$, for example, the k value for a specimen

saturated with deionized water was about 14% of that saturated in sodium chloride (NaCl) solution, and it was about 7% in calcium chloride (CaCl_2) solution. Under otherwise similar conditions, the S_u values of the specimens were consistently greater in deionized water than that in saline solution, with less significant differences between the two types of saline solution. The above observations suggest that pore-fluid salinity and composition both play important roles in determining the engineering properties of soil-bentonite mixtures, particularly hydraulic conductivity. However, the differences in k and S_u are considered relatively negligible by varying the concentration of each saline solution (i.e. 2%, 5%, and 10%).

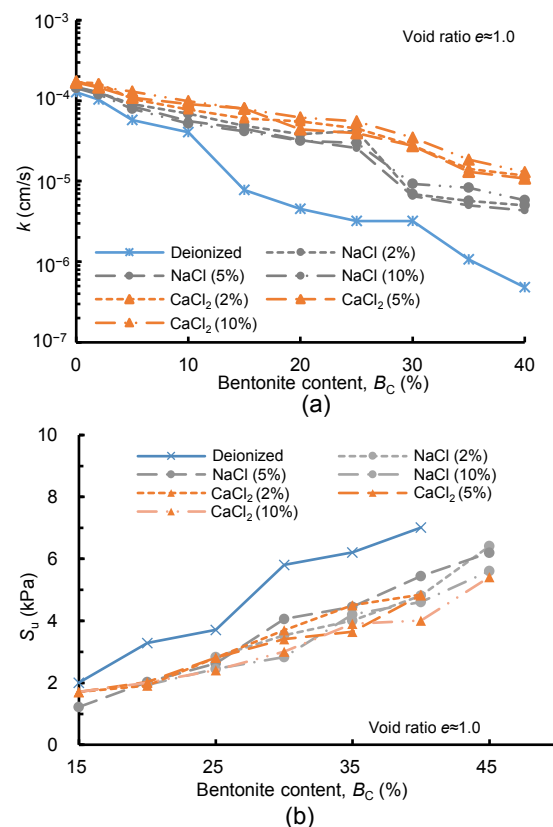


Fig. 1 Hydro-mechanical properties of soil-bentonite mixtures: (a) hydraulic conductivity (k); (b) undrained shear strength (S_u)

2.2 Microscale considerations

The microstructural characterization of soils is gaining increasing prominence in finding the underlying mechanism of the above experimental observations. In the context of microscale analysis, environmental scanning electron microscopy (ESEM) was

adopted in the current study. It has the advantage of examining wet soil samples under controlled environmental conditions, so that the natural soil characteristics are well preserved (Xue and Lu, 2008; Shang et al., 2015; Lan et al., 2019; Liu et al., 2020a). Microscale images taken at 5000 \times magnification under different conditions are presented in Fig. 2.

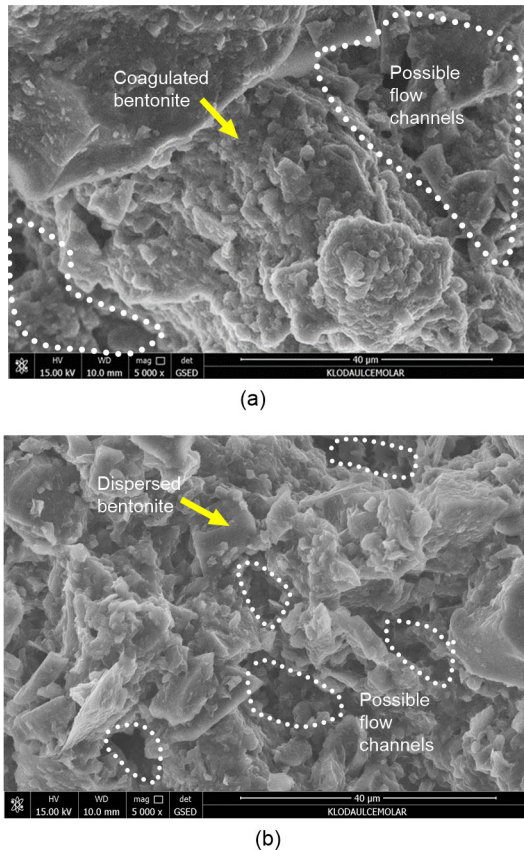


Fig. 2 SEM images of soil-bentonite mixtures ($B_C=20\%$) and schematic illustrations in CaCl_2 solution (10%) (a) and deionized water (b)

Despite having the same bentonite content, clay particles in deionized water (Fig. 2b) are clearly less coagulated than those in saline solution (Fig. 2a), the latter having a massive structure with less discernable flow channels. The potential reason is the different contribution of diffused double layer of the swelling clay. In brief, the rise of ion concentration in the saline solution results in the compression of diffused double layer of the bentonite, thus yielding more opened flow channels in the mixtures with mostly parallel arrangements of clay particles formed by van der Waals attraction (Fig. 2a) (Mitchell, 1993; Mishra et al.,

2015). Moreover, the contact strength formed by van der Waals attraction is much smaller compared to mixtures in deionized water, where the double layers on the clay particles are well developed (Fig. 2b) (Dolinar and Trauner, 2007). This may also explain the lower undrained shear strength of soil-bentonite mixtures in the saline solution.

Furthermore, changes in the connectivity of pores strongly indicate the differences in macro scale behaviors, among which hydraulic conductivity is considered as the fundamental parameter reflecting pore size distribution. In this regard, the pore size distribution curves of soil-bentonite mixtures obtained by magnetic resonance imaging (MRI) are illustrated in Fig. 3. An overall leftward shift of the distribution curves with increasing bentonite content (Fig. 3a) is clearly observed, with the associated mean pore size gradually decreasing. A similar trend was also obtained with a decreased saline solution (i.e. NaCl solution) concentration (Fig. 3b). More specifically, a marked difference is shown between the mixtures saturated with the deionized water and NaCl solution at 2% concentration, while a further rise of the salt concentration results in no substantial differences. Given that the pore size distribution reveals the number of open pores that allows the pore-fluid to flow (Kawaragi et al., 2009; Romero, 2013), this variable is likely to account for variations in the hydraulic conductivity of soils. Compared to results in Fig. 1a, observations on pore size distribution were verified, and they are consistent with the changes of k measured under different conditions.

2.3 Empirical models

The presence of bentonite in the mixtures and the associated response to changes in pore fluid salinity both play an essential role in determining the k and S_u . In the above context, it is of interest to examine whether existing models are still applicable for the soil-bentonite mixtures in the saline solution. A trial is performed herein by adopting the general form of Eq. (1). The performance of this correlation is evaluated using test results of the current study. As shown in Fig. 4, the k values of mixtures (i.e. $B_C=0$ to 45%) are plotted against the generalized stress state (e/e_L). For each type of pore fluid, the best-fit curve with a high coefficient of determination is derived (i.e. $R^2=0.81-0.95$). When test results are fitted in a whole

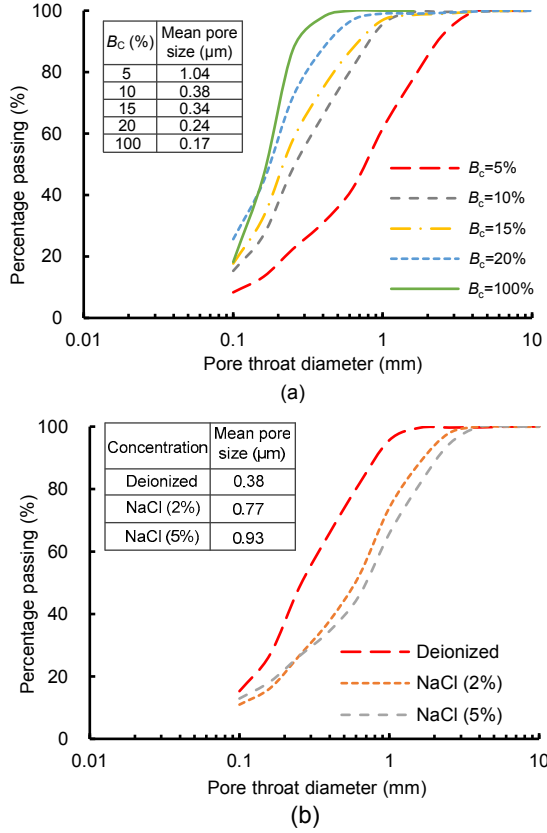


Fig. 3 Pore size distribution curves of soil-bentonite mixtures

(a) With different bentonite contents and in deionized water; (b) With $B_c=10\%$ and in NaCl solution

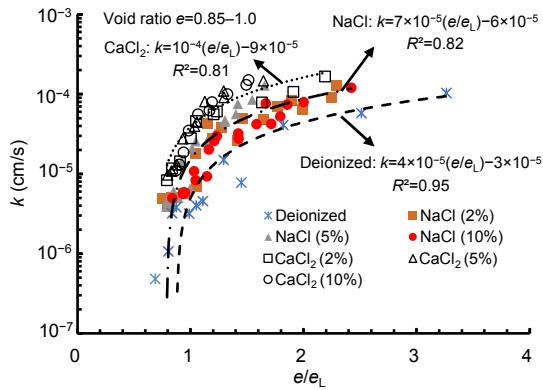


Fig. 4 Relationship between the ratio e/e_L and the hydraulic conductivity k

category with a single best-fit curve, however, the coefficient of determination reduces to 0.53, presumably because the effects of pore-fluid salinity on k are not properly addressed in Eq. (1). Therefore, fur-

ther enhancement via the unified characterization of the hydro-mechanical properties of the soil-bentonite mixtures is needed.

It is worth noting that bentonite clay is considered more sensitive to the varied conditions of pore-fluid than crushed silica. Hence, the contribution of bentonite is dominant in evaluating the changes of k and S_u in a mixture exposed to pore-fluid salinity. In this connection, the generalized stress state in Eq. (1) is modified, and it specifies a new term as e_b/e_L , where e_b is defined as the ratio between void volume and bentonite volume. Alternatively, e_b can be derived in the following form:

$$e_b = e \cdot \frac{G_{sb}}{G_s \cdot B_c}, \quad (2)$$

where G_{sb} and G_s are the specific gravities of bentonite and the mixture, respectively. Assuming that the bentonite clay is fully saturated at the liquid limit ($S_r=1$) and the soil-bentonite mixtures were tested under conditions where S_r is approaching a unit, the following equations can be specified:

$$\begin{cases} e = \frac{G_s \cdot W_C}{S_r} = G_s \cdot W_C, \\ e_L = G_{sb} \cdot LL, \end{cases} \quad (3)$$

where LL (%) denotes the liquid limit of the bentonite (Table 1), whereas W_C (%) indicates the water content.

Table 1 Liquid limit of bentonite clay under varied salinities

Salt content by mass (%)	Liquid limit (%)	
	NaCl solution	CaCl ₂ solution
0	127	127
2	93	85
5	82	74
10	73	65

By combining Eqs. (2) and (3), the new term (e_b/e_L) can be further derived as follows:

$$\frac{e_b}{e_L} = \frac{G_s \cdot W_C \cdot G_{sb}}{G_s \cdot B_c \cdot G_{sb} \cdot LL} = \frac{W_C}{B_c \cdot LL}. \quad (4)$$

The dependence of k is readily established by using the e_b/e_L as a state variable. As shown in Fig. 5a, it is encouraging to see that a unified characterization of k in both deionized water and saline solution is satisfactorily achieved. The general expression for characterizing k is given as

$$k = \alpha_1 \ln\left(\frac{e_b}{e_L}\right) + \beta_1, \quad (5)$$

where the parameters are assigned the following values: $\alpha_1=4\times 10^{-5}$ cm/s; $\beta_1=4\times 10^{-6}$ cm/s. The performance of the proposed model was also examined by using data collected from the literature. As shown in Fig. 5b, measured data on hydraulic conductivity was compared with the predicted values. On average, the predicted ones are within two times of the k value measured. Given that the hydraulic conductivity of soils is often written in logarithmic form, the discrepancies in this plot are negligible. Hence, it is considered with great confidence that the proposed model is going to have a wide range of applications.

Note that the proposed stress state term (e_b/e_L) is anchored with the liquid limit, which examines the water content of a material at particular shear strength; one may be concerned about this new term in Eq. (4), as to whether it applies to empirical correlations involving shearing properties of soils. With this in mind, the undrained shear strength of the mixtures was plotted as a function of the proposed stress state term in Fig. 6. Consistently with our expectations, a fairly good correlation is achieved and the empirical relationship is given as follows:

$$S_u = \alpha_2 \left(\frac{e_b}{e_L}\right)^{\beta_2}, \quad (6)$$

where $\alpha_2=5.59$ kPa and $\beta_2=-1.29$ without unit. Given that Eqs. (5) and (6) are both developed in a sound framework, the new empirical models are beneficial for practical purposes in preliminary design calculations, and to potentially provide an order-of-magnitude check against experimentally determined values. It is also worth noting that the bentonite content and the mass concentration of salts are both set in a limited range in this work. Further research along this line would be certainly worth conducting.

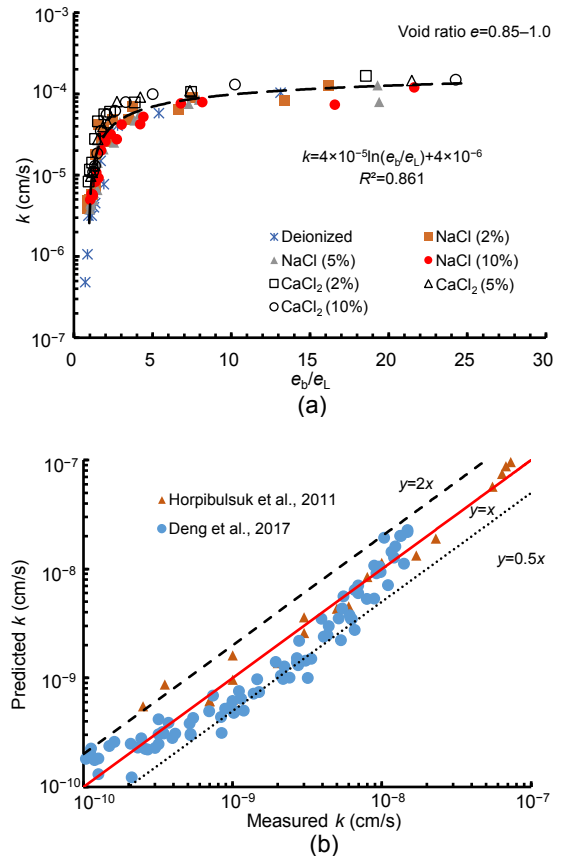


Fig. 5 Proposed model and the associated validation (a) Relationship between the e_b/e_L ratio and hydraulic conductivity; (b) Model validation using data from the literature

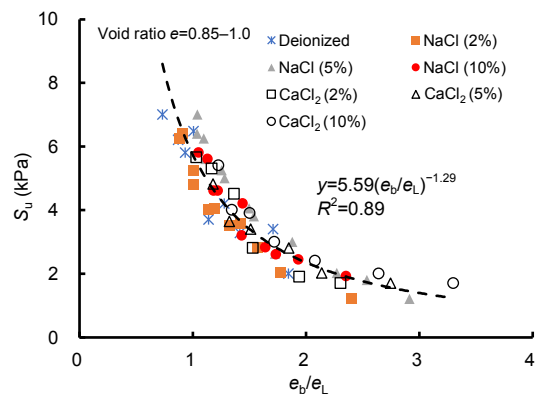


Fig. 6 Relationship between the e_b/e_L ratio and the undrained shear strength (S_u)

3 Conclusions

The main findings of this study are summarized as follows:

1. Values of k declined and S_u increased with the bentonite content in soil-bentonite mixture, with changes more significant in deionized water than those in saline solution. Under otherwise similar conditions, k was greater for the specimens saturated in CaCl_2 solution than for those in NaCl solution.

2. Results of the MRI test indicate that the differences of the pore size distribution are consistent with changes of k .

3. A unified characterization of k and S_u for the soil-bentonite mixtures is established while considering the effect of pore-fluid salinity.

Contributors

Wen-zhe ZHANG and Xiao WEI processed the corresponding data. Xin LIU wrote the first draft of the manuscript. Heng-xing LAN revised and edited the final version.

Conflict of interest

Xin LIU, Wen-zhe ZHANG, Xiao WEI, and Heng-xing LAN declare that they have no conflict of interest.

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List of electronic supplementary materials

Data S1 Unified characterization of hydro-mechanical properties of soil-bentonite mixtures exposed to pore-fluid salinity