Insights to the micro-macro transfer phenomena in clayey soil under electrical gradient

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Abstract
The application of electrical energy during Electrokinetic Treatment (EKT) on soils has been well experimented on vast amount of soils, to either consolidate or decontaminate them. The design of EKT benefits from a deeper knowledge on the phenomena occurring during the treatment, including the settlements prediction. Within this context, this paper analyses the increment of secondary deformations observed in consolidated kaolin slurries when EKT is applied under three different vertical stresses. This is an electro-hydro-mechanical coupled problem because the deformations observed under constant electrical field are stress dependent. The analysis considers the mobilization of the water (adsorbed) at the level of microstructure of the soil by electrical gradient, and therefore volume changes occur when water is flowing from the micro(pores of the clay aggregates) to the macropores (pores between these aggregates).

Keywords: Secondary consolidation, Micro-macro transfer, Clayey soil, Electrokinetic treatment.

Introduction
The application of electrical energy during Electrokinetic Treatment (EKT) on soils has been well experimented on vast amount of soils with different conditions, to either consolidate (by dewatering) or decontaminate them ([1], [2], [3], [4], [5]). The design of EKT benefits from a deeper knowledge on the phenomena occurring during the treatment, including settlements prediction. The prediction of settlements during EKT requires solving an electro-hydro-mechanical coupled problem. While there are several solutions for this problem ([6], [7], [8], [9]), as far as the authors know there are no references in the literature to how electrical current affects secondary consolidation. For this reason, the effect of electrical potential on creep deformations was investigated based on the mobilization of water adsorbed in the surface of the electrically charged clay minerals. In addition, it is considered that the temperature is constant and the effects of electrophoresis, electrical gradients caused by ions movement and water electrolysis are negligible. Secondary consolidation in clays has been proposed to be dependent on many factors like the rearrangement of the soil mineral particles and sets of clay particles, mineralogical composition, consolidation pressure, load increments, load history, etc ([10], [11], [12]). Some theories consider soil particle rearrangement under mechanical loading, while others explain the phenomenon of the secondary deformation as the decrease in size of the micro and macro pores of the soil, which comprises of the intra-aggregate and inter-aggregate voids made up of the clay minerals (see Figure 1). It was suggested that the local dehydration process may be approximated by micro consolidation phenomena, in which the clay aggregates lose water toward the larger pores [13].

Figure 1: Micro and micropores in consolidated clay slurries.

In this paper, the above theory is extended to analyze secondary consolidation under the electrical potential dragging the water adsorbed by the clay minerals, besides that from the smaller pores (micropores). Consolidated slurry samples were tested instead of compacted samples, so that the phenomena could be investigated in a more realistic manner, as EKT is been applied to improve soft clayey soils rather than in compacted ones.

Oedometer tests were performed in modified cells to quantify and analyze the electrical effects on secondary deformations measured during constant vertical stresses. Constant electrical voltage was applied only during the secondary consolidation phase, to isolate its effects from those due to primary consolidation. This effect was investigated in different tests, in which EKT was applied in three different vertical stresses, applied to evaluate their effect on this water transfer.
mechanism. The same stress paths were applied in tests performed without electrical action, for comparison.

**Theory**

**Water movement due to EKT**

During EKT, percolation is caused by voltage gradients and by hydraulic gradients, generated by water movement and its accumulation in impervious boundaries [1]. The total flow of fluid extracted can be computed considering the superimposition of hydraulic and electrical gradients, assuming that Darcy's and Ohm's law are valid. Under changes in vertical stresses hydrodynamic consolidation occurs (Terzaghi theory) because volume changes are caused by water expulsion from the pores of the saturated soil. When EKT is applied, the flow of fluid due to the hydraulic and electrical gradients are superimposed to obtain the total flow. The hydraulically driven flow $Q_h$ is in progress mainly while pore pressures generated by the stress increment are dissipating (primary consolidation). This flow is described by Eqn. 1, where $k_h$ is the saturated permeability (or coefficient of hydraulic conductivity) (in m/s), $A$ is the cross-section area perpendicular to flow and $i$ is the hydraulic gradient given by the change in water head $H$ along the direction of flow $x$, caused by changes in pore water pressure.

$$Q_h = -k_h i A = -k_h \frac{dH}{dx} A \quad (1)$$

Settlements rate decreases in asymptotic manner due to the decrease in hydraulic gradient, permeability and soil compressibility. It can be assumed that soil volume changes are due to the reduction of the sizes of the pores, to which the biggest pores (macropores) will have the largest contribution. Such is illustrated in Figure 2 (from a to b in the plot, b configuration). These changes are considered to be at the level of the macrostructure and usually are assumed to mark the completion of 90% of primary consolidation. It comes natural to assume that the volume changes in secondary consolidation are at the level of the smallest pores, the micropores (from b to c in the plot in Figure 2, c configuration). Therefore, secondary deformations are caused by a delayed response of microstructure to stress changes. This is equivalent to say that changes in effective stresses are delayed in the microstructural level. This idea is included in some descriptions of delayed deformations and in some theoretical formulations ([13], [14]).

Both bulk water and the water adsorbed are mobilized by the electrical potential. When comparing the hydraulically and the electrically driven water flows, the main difference is that water and ions adsorbed to the clay minerals and in the double layer can be mobilized by electrical current. The exchangeable positively charged ions tend to migrate towards the cathode, thus dragging the adsorbed water from the clay surface along with it. The proportion of adsorbed water and ions depends on the specific surface, chemical forces and electrostatic forces on the clay mineral. Forces like double layer repulsion and van der Waals attraction hold the clay particles in various fabric formations. These forces can be affected by pH changes and therefore by electrical current. External stress also affects fabric as they affect the density of the soil, and therefore the proximity of the clay aggregates and the amount of bridges between the clay minerals through which current can flow [15]. This concept is used in this paper to explain creep observed when electrical current is applied.

![Figure 2: Water and ions exchange during consolidation under mechanical and electrical potential for kaolin](image)

Depending on the clay minerals, the contribution of the adsorbed water to electrical current flow can be larger than that of the bulk water [16]. In the formulation of the mass transfer process using Gibbs free energy from the classical thermodynamics of soil water, the exchangeable cations are assumed to be a part of soil skeleton. If the ions are electrically forced out from the double layer, the mass of water which is balancing these ions will also move with them, thus decreasing the overall volume of the soil minerals. This reduces the repulsion forces between the clay minerals and it can be expected that deformations will occur due to changes in the electrical potential of this water (micro-water). Such effect is also illustrated in Figure 2 (b to c’ in the plot, c’ configuration).

Because, in this paper, EKT was applied after 90% of primary consolidation, the change in the overall volume caused by the reduction in micro pores will be visible from the macroscopic point of view only during the secondary consolidation phase. However, the resulting arrangement may not be very important for global volume changes due to the less impact of individual microstructural changes on macrostructural behaviour [17]. The electrical force can also affect the reversibility of the volume change processes from a macrostructural point of view, but this analysis is out of the scope of this paper.
Being total void ratio, e, the sum of the void ratio of the macropores e_{m} (volume of macropores to the volume of solids) and of the micropores, e_{m} (volume of micropores to the volume of solids), basically changes in e_{m} are dominant in primary consolidation and mainly changes in e_{m} occur in secondary consolidation. In the case of consolidated slurry clays, water will flow from the micropores to the macropores. While for compacted clays the distinction between macropores and micropores is done considering to be, respectively, the pores between the clay aggregates and in the clay aggregates, for natural soft clays and remolded or in consolidated slurry materials (in this work they were prepared with water content 1.5 times the liquid limit, as suggested in [18]) this is not evident. Nevertheless, the formation of aggregates occurs naturally from the suspension and consolidation process and therefore the consideration of sets of clay particles, named here as aggregates of clay minerals, remains valid. For these materials, it will be assumed in this paper that the macropores are the pores where water is free and the micropores are those where water has reduced mobility (immobile water) (see Fig. 1). This water will be named as micro water.

The model proposed by Navarro & Alonso [13] explains secondary consolidation resulting from water transfer from the micro to macro pores caused by a thermodynamic force, which exists due to chemical imbalance of water. This transfer is at a very slow rate and continues to reduce the void ratio (micropores contribution) until reaching equilibrium. Volume reduction of the microvoids is complete when the change in effective stresses in the microstructure is equal to stress change applied to the soil. This model will be extended to the case where electrical potential is applied during the secondary consolidation phase. Both are formulated under the hypotheses of ideal fluids in both micro and macropores, water structure is the same in the two types of pores, exchangeable cations are at the level of the mineral phase and will not contribute for this balance, and the effect of dissolved solutes is ignored. Temperature is assumed constant.

### Hydrodynamic Secondary Consolidation

According to Navarro & Alonso [13], there is water transfer between the micro and the macrostructure while the chemical potential of the water existing in both levels is not in equilibrium considering Thermodynamic. The mass exchange of water, c_{m} is given by Eqn. 2 where \( \pi \) and \( \pi_{b} \) are the pressure acting in the microstructure and the swelling pressure (the pressure necessary to mobilize the microstructure). This equation means that there is water transfer while the pressure applied in the microstructure is larger than swelling pressure, the pressure below which there is no volume change of the microstructure. Parameter G describes the speed of water transfer between the micro and the macro pores and is assumed to be constant along time. It depends on initial constant \( G_{0}/\rho_{m0} \) and, through parameter C, on the permeability, compressibility and size of the clay clusters retaining the micro water (Eqn. 3). These parameters can be calibrated with experimental data. D is the parameter obtained directly from the consolidation tests and is equivalent to a compressibility index but at the level of the microstructure, valid for that clay under changes in effective stress. This equation is valid only after the end of primary consolidation, assuming that all changes in void ratio measured in the tests are changes in the microstructural void ratio. Total void ratio e in Eqn. 4 is assumed to be void ratio at the end of primary consolidation phase (point b in Figure 2).

\[
\frac{\Delta e_{m}(t)}{\rho_{m}} = \frac{G_{0}}{\rho_{m0}} e^{-\Delta e_{m}/C}
\]

\[
\Delta e_{m}(t) = D \ln \left[ \frac{\sigma_{i} / \sigma_{i-1}}{1 + (\sigma_{i} / \sigma_{i-1}) \exp \left[ (1 + e) \frac{G_{0} \, \pi_{b}}{\rho_{m} \, D} \right]} \right]
\]

The electrical potential can now be introduced in this water balance, described in next section.

### Effect of Electrical Potential

Pore pressure induced by EKT is named as \( u_{EK} \) (Eqn. 5) and can be computed considering the generation of an opposite hydraulic flow, \( Q_{h} \) (Eqn. 1) [1]. For the hydraulic gradient, it is considered \( H = h/u_{y_{w}} \), where \( u_{y_{w}} \) is the water volumetric weight (assumed \( \gamma_{w} = 10kN/m^{3} \)). \( u_{EK} \) is considered constant while voltage gradient is constant, however this is valid only if both \( k_{a} \) and \( k_{h} \) remain unchanged during the application of the electrical potential. This can only be accepted in secondary consolidation, where changes in void ratio are very small.

\[
du_{EK} = \frac{k_{c} \, dy_{w}}{k_{h} \, dx}
\]

For the tests performed it can be assumed that only microwater is mobilized by electrical potential because the electrical potential was applied after the completion of primary consolidation. In this case, only the definition of the mass-transfer rate per unit volume, \( c_{m} \) (Eqn. 2) changes because an additional pressure, \( u_{EK} \), is acting in the microstructure. The mass-transfer rate per unit volume considering EKT is now named \( c_{m, EK} \) (Eqn. 6).

\[
c_{m, EK} \approx G (\pi - \pi_{b} + u_{EK}) = c_{m} + G u_{EK}
\]

From a physical point of view, the changes in volume motivated by the electrical gradient will affect parameter G and D (further details in [19]). If D is not considered as an intrinsic soil parameter, the changes in its value must have to be considered if further loading steps are applied because of the contractile activity of electrical potential in this type of soil. The relevance of this correction will be investigated in future work.

The effect of the new ‘boundary’ condition, \( u_{EK} \) means that less stress is necessary to mobilize microwater flow when electrical potential is applied. This result was expected, as it is one of the reasons why electrokinetic treatment is adopted. The changes in microstructural void ratio will be estimated considering the validity of Eqn. 4, also because it is difficult to determine \( u_{EK} \) as it requires knowing \( k_{a} \) and \( k_{h} \) for the current void ratio. Therefore, the changes in \( G/\rho_{m} \) (or \( G_{0}/\rho_{m0} \) and C) and in D will be estimated from the experimental data. This is due to the reduction of the repulsion forces in the microstructure, already discussed, reducing the size of the
micro pores. This size reduction is interpreted as an increase in the compressibility of the microstructure.  

The simplifications introduced allow understanding the increase in volume changes experienced by clayey soils under the influence of electrical potential, and consequent increment observed for the coefficient of secondary consolidation $C_a$:

$$c_m = \frac{G(a - \pi_a)}{\Delta \log t}$$  

This coefficient is the variation of the void ratio $\Delta e$ over the time $t$ starting after the end of the primary consolidation and is defined by Eqn. 7 [12], where $\Delta e$ is assumed to be equal to $\Delta e_m$.

$$C_a = \frac{\Delta \log t}{\Delta e_m}$$  

Material and Experimental Tests

Commercially white kaolin clay was adopted in this experimental research aiming to simplify data treatment because it is a Portuguese non-expansive clayey soil. The material was supplied in powder, with 68% in mass of grains with silt size (diameters between 0.075 mm and 0.002 mm) and 31% with clay size (diameters smaller than 0.002 mm). Solid volumetric weight is 26.1 kN/m$^3$. Liquid limit is 52%, and plasticity index is 22%, therefore the material classifies as highly plastic silt (MH) according to the Unified Soil Classification System. The zeta potential value (ZP) for this material was supplied in powder, with 68% in mass of grains because it is a Portuguese non-expansive clayey soil. The experimental research aiming to simplify data treatment was performed in stages up to 12 kPa to consolidate the soil. The anode and cathode were at the bottom and top of the sample respectively. The DC voltage/current source Aim-TTi EX354RD Bench Power Supply, 280W (0-35 V and 0-4 A) was used. Silver plate electrodes were used (area 0.0004 m$^2$).

![Electrokinetic test apparatus](image)

Settlements due to secondary consolidation were measured in independent tests performed for each vertical stress. The stresses 50, 200 and 800 kPa were investigated. One set of tests was performed without electrical current (control tests), while electrical current was applied in the other set (EK tests). A total of 6 tests were performed. The tests details are shown in Table 1. The first are the control tests which are used for comparison purpose. The DC Voltage of 2V was applied 24 hrs after loading the sample with the desired stress in order not to overlap with the primary consolidation phase.

<table>
<thead>
<tr>
<th>Test</th>
<th>Control</th>
<th>EKT</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-D Oedometer</td>
<td>1</td>
<td>2</td>
</tr>
<tr>
<td>Void ratio*</td>
<td>1.136</td>
<td>0.917</td>
</tr>
<tr>
<td>Stress (kPa)</td>
<td>50</td>
<td>200</td>
</tr>
</tbody>
</table>

* at the beginning of the loading interval under investigation

Loading was applied as per ASTM-D2435 [22]. All the loading stages prior to the final load stage where EKT was applied, were lasted for 24 hours. A total time of 24 hrs was given for the final loading before the period under analysis, which were the following 3 days (72 hours) under the same loading, both for the control and EK tests (24h on-24h off-24h on, 3 days).

Results

![Electrokinetic test apparatus](image)
**Electrical anisotropy**

Table 2 presents the values of the net conductivity $\sigma$ measured for the samples having the different particle orientations mentioned. For increasing stresses, the conductivity of the soil decreased in all the cases. As expected [20], the value found for the parallel flow samples are greater than the perpendicular ones and, for both orientations, the conductivities decrease with increasing vertical stress. This may be explained by some water expulsion during compression and confirms the electro-hydro-mechanical coupled behaviour of the clays.

The relationship between the two values ($\sigma_{\text{prl}}$/$\sigma_{\text{pnd}}$) is the coefficient of electrical anisotropy. Since the electrical flow will be more in parallel flow samples, the values are larger than unity (Table 2). The fact that electrical anisotropy was found for the kaolin samples proves that this non-active clay is able to transmit electrical current through its minerals, and therefore validates the hypothesis adopted that the increment of secondary consolidation to be caused by the mobilization of water in the clay minerals.

**Secondary consolidation**

The analysis of the behaviour along time was performed by studying the changes in coefficients of secondary consolidation (Eqn. 7) and in the microstructural parameters. The values found for the coefficient of secondary consolidation for the loading steps of the two sets of tests are presented in Figure 4 ($C_u$ refers to the control tests and $C_{\text{dek}}$ to EK tests). They were computed considering the total duration of the secondary consolidation period (3 days).

![Figure 4: Coefficient of Secondary Consolidation for kaolin](image)

The points in Figure 4 were adjusted to highlight the differences of the two cases. It can be seen that $C_u$ decreases for increasing vertical stress, while $C_{\text{dek}}$ remains approximately constant (around 0.009). For two stresses, the values of $C_{\text{dek}}$ are higher than those of $C_u$ and factor $C_{\text{dek}}/C_u$ is larger than one. This result clearly shows that the amount of water expelled from the micro pores due to the electrical potential is greater than that of the control samples and confirms that electrokinetic flow increases water flow to influence the secondary consolidation. This domain is expected because pore pressures generated by stress increment in that loading step are null after primary consolidation (already discussed), and the only pore pressure is caused by electrokinetic flow.

<table>
<thead>
<tr>
<th>Stress (kPa)</th>
<th>$\sigma_{\text{pnd}}$ (S/m)</th>
<th>$\sigma_{\text{prl}}$ (S/m)</th>
<th>$\sigma_{\text{prl}}$/$\sigma_{\text{pnd}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>50</td>
<td>0.052</td>
<td>0.075</td>
<td>1.43</td>
</tr>
<tr>
<td>200</td>
<td>0.039</td>
<td>0.060</td>
<td>1.91</td>
</tr>
<tr>
<td>800</td>
<td>0.028</td>
<td>0.046</td>
<td>1.78</td>
</tr>
</tbody>
</table>

* perpendicular flow  ** parallel flow

The constants for the model were calibrated using the conventional consolidation test data as shown in Table 3. They are the micro-macro transfer coefficient ($G/\rho m$) and its variation with secondary deformation (C). The analysis was done considering the three days of secondary consolidation. The changes in $e_m$ (given by $e_{\text{init}}-e_m$) were assumed to be equal to the changes in total void ratio during secondary consolidation (Figure 5). This is valid under the hypothesis that secondary displacements would be caused only by microstructural arrangements. Accordingly, in all load steps, the value of $e_{\text{init}}$ was considered to be the void ratio at the beginning of that loading step minus the void ratio at the beginning of secondary consolidation (3 days).

![Table 3: Parameters for the transfer rate coefficient G](image)

**Table 3: Parameters for the transfer rate coefficient G**

<table>
<thead>
<tr>
<th>Stress (kPa)</th>
<th>$G/\rho m$ (s.kPa)</th>
<th>$C$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>EK</td>
<td></td>
</tr>
<tr>
<td>50</td>
<td>2.0E-07</td>
<td>0.0024</td>
</tr>
<tr>
<td>200</td>
<td>2.0E-08</td>
<td>0.0019</td>
</tr>
<tr>
<td>800</td>
<td>2.0E-08</td>
<td>0.0021</td>
</tr>
</tbody>
</table>

**Figure 4**: Coefficient of Secondary Consolidation for kaolin
The authors acknowledge the ERASMUS MUNDUS Action2 Strand 1, EACEA/42/11 (Grant Agreement 2012-2649/001) for the financial support and FCT, Portugal for funding the experimental tests (EXPL/ECM-GEO/0109/2013).

References


Conclusions

The analysis allowed to find some conclusions, which contribute to understand the Electro-hydro-mechanical phenomena occurring during EKT.

The results of parameter C also indicate that the microstructure compressibility changes more along time when electrical potential is applied. The increase in C also reflects the drag of microstructural water due to electrical potential.

Acknowledgements

Figure 5: Comparison of the control and ek curves for vertical stress a) 50 kPa; b) 200 kPa and b) 800 kPa defined to calibrate parameter $G/\rho_m$ (s.kPa)$^{-3}$

In both the cases, the value of $G/\rho_m$ was found to be decreasing exponentially with the secondary deformations. The $G/\rho_m$ value decreases with increasing vertical stress, or with decreasing void ratio at the beginning of the secondary consolidation, which means that water loses mobility due to micro-porosity reduction. Larger values are observed for the control tests than for the EK tests because, in the latter, the water is mobilized also by the electrical potential, and therefore flow is not governed only by mechanical processes.

The compressibility of the microstructure increased with EKT. This was explained by the mobilization of the adsorbed water in the clay minerals, reducing the repulsive forces. This drag of water resulted in the increment of the settlements speed given indirectly by parameter $G$.

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