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Influence of pore water chemistry on hydraulic conductivity of kaolinite suspensions

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Abstract

The role of pore water chemistry on particle aggregation and soil formation during sedimentation process has been investigated for a kaolinite clay suspensions. The modification of surfaces charge properties of clay minerals induced by different pore water chemistry rules the particles arrangement. As a consequence, at volume scale the sedimentation behaviour and in turn the physical, mechanical and hydraulic behaviour of the formed soil skeleton are changed. Sedimentation tests on several suspensions showed the influence of different aggregation on settling and self-weight consolidation stages. Hydraulic conductivity of kaolin suspensions has been estimated from the settling velocity recorded in sedimentation tests. Test results showed the relevant dependency of the hydraulic conductivity of the clay suspensions on the particle arrangement induced by the pore water chemistry.

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1. Introduction

The influence of pore water chemistry on particles aggregation process is a key issue for understanding the soil skeleton formation and its effect on the macroscopic behaviour of soils. The modification of surface charge properties of clay minerals together with ionic valence and concentration of pore water rules the aggregation process. The ions dissolved in the clay water system tend to neutralize the charge on the edge and face sites of particles surface, controlling the chemo-physical interactions and the particles arrangement [1]. The interparticle forces (Coulombic forces, double layer repulsions, Van der Waals attractions) promote different type of particle associations: edge-to-face (EF) flocculation, edge-to-edge (EE) flocculation, face-to-face (FF) aggregation, and dispersed-deflocculated fabric [2]. The sedimentation test is a useful tool for understanding the effect of the pore water chemistry on the microstructural features of clay-water systems. Two typical curves can be identified (Fig. 1) from sedimentation tests, depending on the degree of particles association in the suspension. Figure 1a shows the settling of a flocculated system, with the suspension height progressively reduced over time leaving a clear supernatant fluid. Figure 1b shows a dispersed system maintaining its configuration over time. In a flocculated system three stages of the sedimentation process can be identified (Fig. 2). During the initial stage (induction period), the electrochemical interactions [3] induce the aggregation from small clusters to aggregates depending on the surface charge distribution of particles. Its duration is linked to the degree of particles or aggregates association [4]. A slight translation of the interface between suspended solids and water can be appreciated at this stage. The flocculated soil particles settle under gravity forming a deposit of increasing height which undergoes self-weight consolidation. Settling and self-weight consolidation take place simultaneously, as evidenced in Figure 2 with reference to time t^* . The soil formation line (Fig. 2) delimitates the sedimentation and the self-weight consolidation regions and is characterised by a constant value of the void ratio e_m , the soil-formation void ratio, which represents the void ratio characterising the soil skeleton at zero effective stresses [5]. At time t_s , corresponding to the intersection between the soil formation line and the line representing the upper interface of the suspension, an abrupt reduction of settling velocity occurs, indicating the completion of the sedimentation stage [6, 7]. During the subsequent self-weight consolidation stage the sediment reduces its height with a decreasing rate until the final height. The duration t_f of primary consolidation process generally lasts a time interval of two order of magnitudes larger than t_s , time necessary to complete the free settling stage [6,7,8].

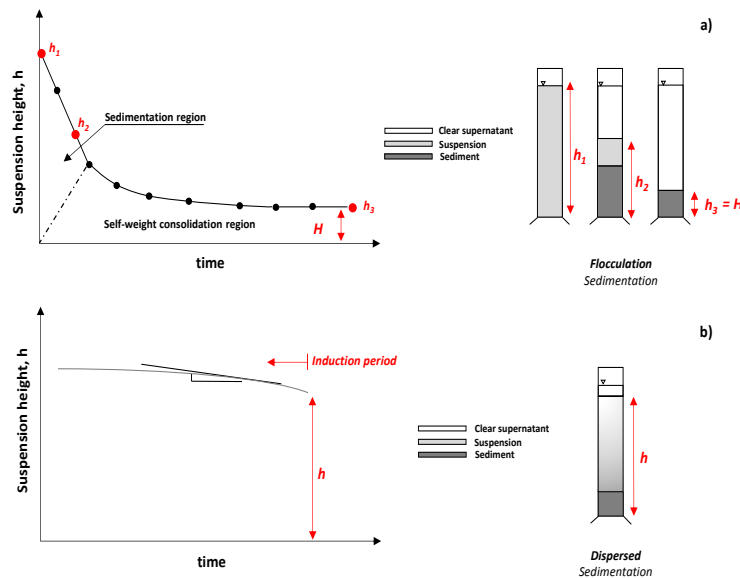


Fig. 1. Sedimentation behaviour of (a) flocculated system; (b) dispersed system (after Vitale et al. [12]).

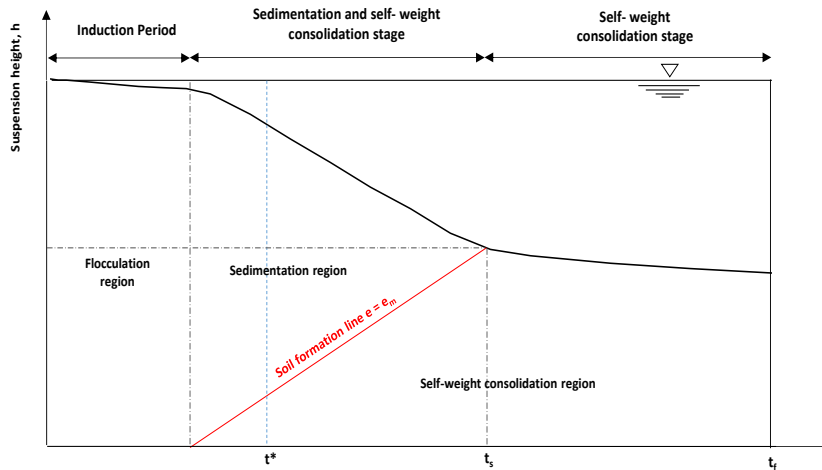


Fig. 2. Stages of sedimentation process (after Vitale et al. [12]).

Sedimentation of kaolin suspensions has been performed and interpreted as function of pH, type of electrolytes and ionic concentration, taking into account the effect of clay particle interaction on the soil skeleton formation. The hydraulic conductivity of suspensions and the formed soil skeleton under zero effective stress have been estimated from the settling velocity recorded in the early stage of the sedimentation test [6]. The experimental evidences at volume scale of the sample were complemented by investigations at particle scale. The soil fabric evolution and its effect on the physical properties of soil have been investigated by means of Dynamic Light Scattering measurements. The experimental results allowed an interpretation in terms of microstructural arrangement of clay particles induced by different pore water chemistry.

2. Materials and procedures

Speswhite kaolin from deposits in the South West of England was the soil considered for the experimental investigation. The specific gravity G_s is 2.6 and its surface area is $14 \text{ m}^2/\text{g}$. The pH value is 4.6, and plastic limit and liquid limit are respectively $w_p = 32\%$ and $w_L = 70\%$, with a plasticity index IP equal to 38%. The mineralogical composition is mainly formed by kaolinite clay minerals with small amounts of quartz and muscovite. No swelling clay phases were detected after the ethylene glycol treatment.

The sedimentation behaviour of kaolin suspension has been investigated as a function of electrolytes concentration (i.e. 1%, 3% and 5% of calcium oxide by dry weight of soil) and source of calcium ions (calcium chloride, CaCl_2 vs. calcium oxide, CaO). Suspensions were prepared in graduated cylinders with mixture of 50g of dry kaolin at 100% of water content. Different amounts of CaO and CaCl_2 were added to reach the required pH and the selected electrolytes concentrations. Additional water was added to get an initial suspension volume of 1000ml. Kaolin suspension treated with CaCl_2 solution was prepared at the concentration of a saturated lime solution ($[\text{Ca}^{2+}] = 22 \text{ mmol/l}$). The pH value was increased by adding KOH. During the test the suspensions heights were monitored as a function of time.

Hydraulic conductivity of clay suspension was determined from the settling velocity of the interface at the early stages of the test according to the equation (1) [6]:

$$K(e) = \frac{v_s(1+e_0)}{\gamma^*} \quad (1)$$

where v_s is the settling velocity of solids, e_0 is the initial void ratio of the suspension and the non-dimensional constant γ^* is given by equation (2)

$$\gamma^* = \frac{(\gamma_s - \gamma_w)}{\gamma_w} \quad (2)$$

The evolution of the average dimension of particles agglomerates over time has been investigated by means of dynamic light scattering (DLS) measurements. The measurements were performed on kaolinite suspension (100mg/l) in deionised water and in presence of $\text{Ca}(\text{OH})_2$ and CaCl_2 solutions at pH 12.4 using a Malvern Nano Zetasizer apparatus with a scattering angle of 173° .

3. Results and discussion

Sedimentation curves of kaolin suspensions prepared at increasing electrolytes concentration, namely 1%, 3% and 5% CaO are reported in Figure 3a. The evolution of the suspension height is shown as a function of the time. In deionised water kaolin suspension shows a flocculated behaviour (pH=4.6). In an acidic environment the mutual interaction between clay particles is affected by the particle surface charge as a result of Coulombic attractions between positively charged edges and negatively charged faces of clay particles, promoting an edge-to-face arrangement which evolves in the short term, as confirmed by DLS measurements reported in Figure 4. A change in the settling characteristics was observed after lime addition. The alkaline environment and divalent cations promoted a more efficient particles association process as revealed by the higher settling velocity of the interface. Increasing amount of lime (from 1% CaO to 3% CaO) promoted higher degree of flocculation of particles, as shown in Figure 3a, with an increase of the settling velocity of the interface and an increase of the final sediment height after the self-weight consolidation stage. No significant modification were observed for samples treated with higher lime percentages (i.e. 5% CaO). The higher degree of particles flocculation is the consequence of the calcium ions adsorption on the negatively charged surfaces of kaolinite particles. The repulsive effect imposed by the modification of pH is counteracted by Van der Waals attractions [2] and an increase of the flocculated aggregates size is detected, as confirmed by DLS measurements results (Fig. 4). The aggregation of particles of lime treated suspensions can be considered not affected by the ongoing pozzolanic reactions since the slow reactivity of kaolinite clay minerals to promote the precipitation of new hydrated phases within the considered time interval [9,10].

The influence of the source of calcium cations on the sedimentation behaviour is shown in Figure 3b. The source of calcium ions in suspension slightly affected the settling velocity of the interface. Compared to the saturated lime solution, a longer settling period and a slight decrease of the final sediment height were observed for CaCl_2 kaolin suspension. DLS measurements showed a lower degree of particles flocculation for samples treated with CaCl_2 solution at pH 12.4 (regulated by KOH addition). Even if the initial concentration of Ca^{2+} is equal, the chemo-physical interactions among particles seems to be affected by the presence of chloride anions, whose adsorption results in a weaker particle association process [11].

The hydraulic conductivity of the kaolin suspensions calculated according to Eq. (1) are reported in Table 1. The evolution of hydraulic conductivity as a function of lime content is shown in Figure 5a. The alkaline environment and the addition of calcium ions increases the hydraulic conductivity. No relevant changes were observed with the addition of lime contents higher than 3% CaO. The particle arrangement markedly influences the hydraulic conductivity of the treated samples, both in the suspended and in the soil-like state, with an increase of permeability of the lime-clay-water mixtures corresponding with an increased average dimensions of treated clay aggregates. The influence of source of calcium ions on the hydraulic conductivity of kaolin suspensions is shown in Figure 5b. Lower values of the hydraulic conductivity were detected for CaCl_2 treated suspensions compared to CaO treated suspensions, as a consequence of the lower degree of aggregation of soil particles, confirmed by the DLS measurements (Figure 4).

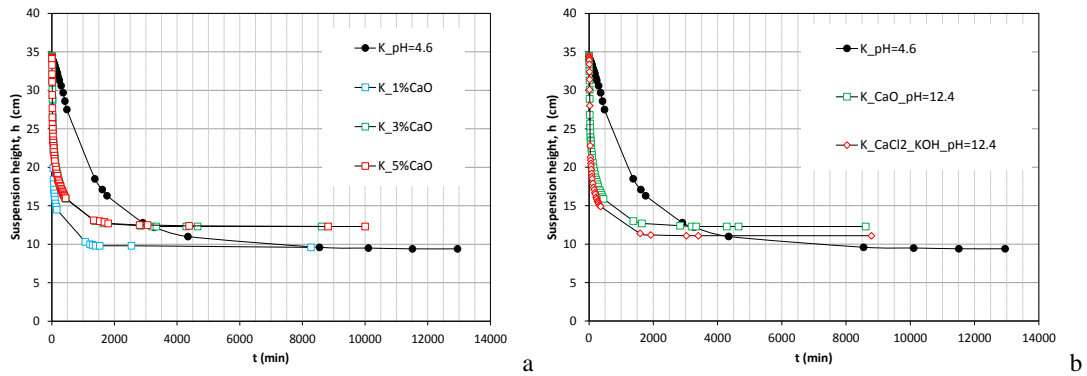


Fig. 3. Sedimentation curves of kaolin suspensions as function of a) lime contents, b) source of calcium ions.

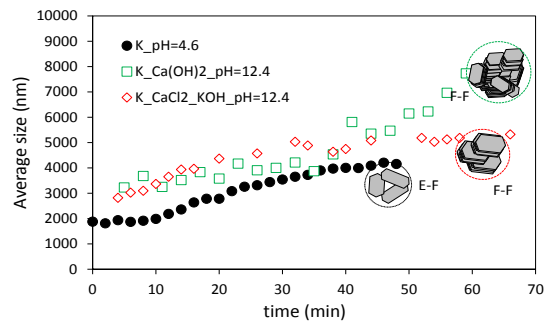


Fig. 4. Evolution of the average aggregates size of kaolin suspensions.

Table 1. Values of k from sedimentation tests on Speswhite kaolin

| Test | e_0 (-) | v_s (m/s) | $K(e_0)$ (m/s) | $K(e_m)$ (m/s) |
|---------------------|--------------|-----------------------|-----------------------|-----------------------|
| K_pH=4.6 | 50 | 2.18×10^{-6} | 6.95×10^{-5} | 3.28×10^{-5} |
| K_1%CaO | 50 | 2.98×10^{-5} | 9.34×10^{-4} | 5.01×10^{-4} |
| K_3%CaO | 50 | 5.07×10^{-5} | 1.60×10^{-3} | 1.24×10^{-3} |
| K_5%CaO | 50 | 5.21×10^{-5} | 1.63×10^{-3} | 1.21×10^{-3} |
| K_CaCl ₂ | 50 | 4.52×10^{-5} | 1.41×10^{-3} | 9.34×10^{-4} |

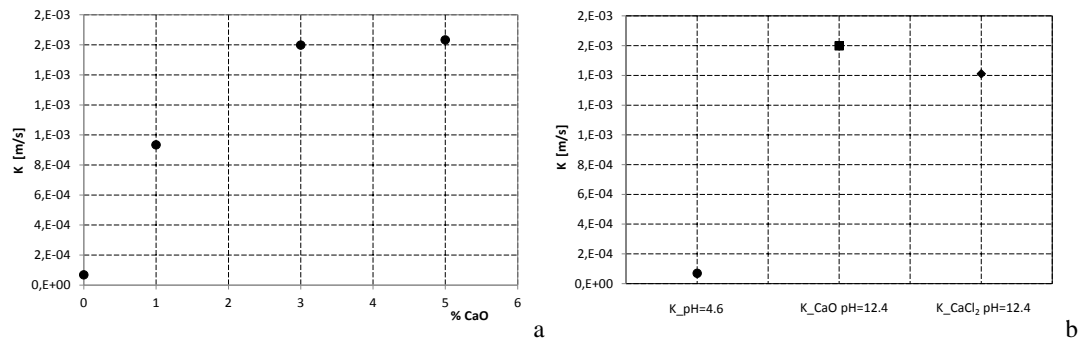


Fig. 5. Hydraulic conductivities at reference void ratio (i.e. e_0) as a function of a) lime contents and b) source of calcium ions.

4. Conclusions

A multi-scale analysis on the sedimentation behaviour of clay suspension under different pore water chemistry highlighted the chemo-physical mechanisms controlling the particle arrangement and the soil skeleton formation.

The aggregation of kaolinite particles is strongly pH dependent. A flocculated behaviour of kaolin suspension in deionised water (pH=4.6) is the consequence of the edge to face arrangement between positively charged edges and negatively charged surfaces. The addition of calcium ions at high pH environment promoted the formation of larger particles aggregates. The effect of aggregation induced by lime is reflected on the sedimentation behaviour of kaolin suspension with a faster settling stage. Similar behaviour was observed for $CaCl_2$ suspension although a less effective particle association process is evidenced by sedimentation test and confirmed by DLS measurements. The physical aggregation of suspended particles induced by pore water chemistry affects the hydraulic conductivity of clay-mineral mixes, with an increase of hydraulic conductivity detected after lime addition. The observed hydraulic behaviour was linked to the different aggregation induced by the pore water chemistry.

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