



Research paper

Influence of hydrated lime on the surface properties and interaction of kaolinite particles

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ABSTRACT

Lime is a widely used chemical additive in the stabilization of problematic soils. However, the physico-chemical mechanism involved, particularly in the short-term, is still not fully understood. In the present work, the influence of hydrated lime ($\text{Ca}(\text{OH})_2$) on the rheological properties of kaolinite dispersion has been investigated. The influence of the type of cation and pH on the interfacial chemistry and particle interaction were also examined. The result showed that kaolinite predominantly adsorbs Ca^{2+} and CaOH^+ at $\text{pH} = 7$ and $\text{pH} = 12.6$ respectively. With increasing concentration of $\text{Ca}(\text{OH})_2$, the value of storage modulus (G'), Bingham yield stress (τ_B) and cohesive energy density (E_c) initially decreased at lower concentration (≤ 5.5 mmol/l) followed by increase at higher concentration (≥ 11 mmol/l); in contrast these parameters decreased monotonously with increasing the concentration of NaOH. The strain hardening characteristic of loss modulus (G'') curve was found to be very sensitive to change in surface chemistry and the associated particle organization. The modification in the mechanical properties of the kaolinite with varying amount of $\text{Ca}(\text{OH})_2$ was attributed to the aggregation microstructure of kaolinite particles. The Ca-ion at higher pH promotes linkage between particles and provides the most efficient way to form dense, tightly packed flocs, which behave as individual coarse grained materials (silt or sand).

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1. Introduction

The soft soils that are usually encountered in construction sites are generally unsuitable for construction works due to their weak mechanical properties and poor workability. Besides, clayey soils exhibit volume change in response to seasonal moisture variation which influences the performance and life time of shallow founded and light weighted structures such as pavements. On the other hand, re-using these materials after stabilization is vital to minimize the cost and environmental impact of construction works. The stabilization of the problematic soils is commonly achieved by mixing them with water and lime or Portland cement and has been thoroughly studied for a long time (Diamond and Kinter, 1965; Choquette et al., 1987; Locat et al., 1990; Verhasselt, 1990; Bell, 1996; Al-Mukhtar et al., 2010; Kavak and Baykal, 2012; Pomakhina et al., 2012; Eisazadeh et al., 2012).

Most of the previous studies cited above refer mainly to the long-term effect of lime on soil property. The physico-chemical mechanism

involved in long-term, i.e. pozzolanic reaction is well understood. In contrast, the mechanism involved in short-term, particularly an increase in unconfined compressive strength (UCS) and shear strength of clayey soil prior to the precipitation of secondary phases (CSH and/or CASH), has not been fully evaluated. This has usually been explained by the reduction of the thickness of diffuse double layer (DDL) around clay mineral particles in the presence of Ca-ion. However, CaCl_2 solution, with similar calcium concentration ($[\text{Ca}^{2+}] = 22$ mmol/l) to that of the saturated lime solution, has no significant effect on mechanical property of expansive clayey materials (Diamond and Kinter, 1965; Verhasselt, 1990). Therefore, the cation exchange reaction and compression of the DDL alone cannot explain the short-term improvement in the mechanical properties of lime treated clayey soil. Understanding the reaction mechanism in short-term is important if one want to optimize the lime treatment technique. In addition, the short-term reaction (first minute up to 24 h after mixing) is a crucial stage as it determines the long-term characteristics (durability and mechanical characteristic) of treated material (Konan et al., 2009).

Surface chemistry determines the interaction between dispersed clay mineral particles (i.e. attractive or repulsive) and affects their probable orientation with respect to each other (Van Olphen, 1964; Kretzschmar et al., 1998; Johnson et al., 1998; Abend and Lagaly, 2000; Penner and Lagaly, 2001; Tombacz and Szekeres, 2004, 2006).

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On the other hand, since the magnitude and direction of the inter-particle forces depend on the separation distance and surface area over which the interaction occurs (Van Olphen, 1964), particle orientation in turn may affect the magnitude of inter-particle cohesive forces and mechanical strength of the aggregated flocs. It is commonly observed that the mode of interactions and resulted floc structures are affected by different parameters such as the ionic strength and pH of the medium (Abend and Lagaly, 2000; Penner and Lagaly, 2001; Tombacz and Szekeres, 2004, 2006; Gupta et al., 2011). Furthermore, pH was indicated to play a role in the hydrolysis reaction of multivalent cation (Mpofu et al., 2003). This is expected to take place during lime addition, since it modifies both the concentration of calcium and pH of the dispersion. Thus, the interfacial chemistry of clay mineral particles and the interaction between them in calcium rich alkaline condition are central in understanding the causes of short-term improvement of the mechanical properties of lime treated clayey soils. In other words, the knowledge of clay mineral particle interactions in the presence of Ca and OH⁻ ions is important.

The rheological behavior of colloidal particles is closely related to the dispersion microstructure at rest and under stress. Consequently, many workers have used rheological techniques to indirectly assess the physicochemical force between colloidal particles and the associated aggregation structure (Johnson et al., 1998; Abend and Lagaly, 2000; Hyun et al., 2002; Mpofu et al., 2003; Tombacz and Szekeres, 2004, 2006; Bossard et al., 2007; Shankar et al., 2010; Chemed et al., 2014). Although the effect of ionic strength and pH on the rheological properties of clay mineral dispersion has been thoroughly studied (Abend and Lagaly, 2000; Duran et al., 2000; Ramos-Tejada et al., 2001; Nasser and James, 2006, 2008; Tombacz and Szekeres, 2004, 2006), a detail description of particle interaction in calcium rich alkaline condition such as in lime solution is rarely presented in the literature. The limited information is considered important, since in addition to determining the aggregation structure and mechanical strength of flocs, it can influence the mixing ability of the soil with the lime solution and affects the long-term reaction.

In the present work, the rheological behavior as a function of hydrated lime was investigated to better understand the role of Ca²⁺ and OH⁻ ions in the modification of surface property and interaction of kaolinite (Kaol) particles. Furthermore, the influence of cation type (Ca²⁺ vs. Na⁺) and pH (Ca(OH)₂ vs. CaCl₂) was also studied. The interfacial chemistry was assessed via calcium adsorption and zeta potential measurements. The size of the aggregated particle structure was estimated using dynamic light scattering (DLS) in order to demonstrate further the particle interaction and flocs formed at different conditions. Finally, the particle aggregation mechanism with varying amount of Ca(OH)₂ is proposed.

2. Materials and methods

2.1. Materials and characterization

Two Kaol samples, KGa-1b (low-defect) and KGa-2 (high-defect) purchased from the Source Clay Repository of the Clay Minerals Society, were used after de-carbonation and Na⁺-homoionization based on the following procedure: carbonates were dissolved at pH = 5 and 80 °C in sodium acetate-acetic acid buffer and Na⁺-homoionization was obtained by three times sodium exchange in 1M NaCl solution followed by several washing and centrifugation cycles until the supernatant was chloride free (silver nitrate test) and had conductivity below 5 μS/cm. Bulk mineralogy of the samples was determined by X-ray powder diffraction (XRD), on randomly oriented samples, with a Bruker AXS D8 device, using CuKα (λ = 0.154 nm) radiation and a step size of 0.021 °2θ. Both samples consist of Kaol (>97 mass%) quartz and anatase (<3 mass% in total). The SEM micrographs confirm the pseudo-hexagonal shape and platy features of the Kaol particles in the two samples (Fig. 1).

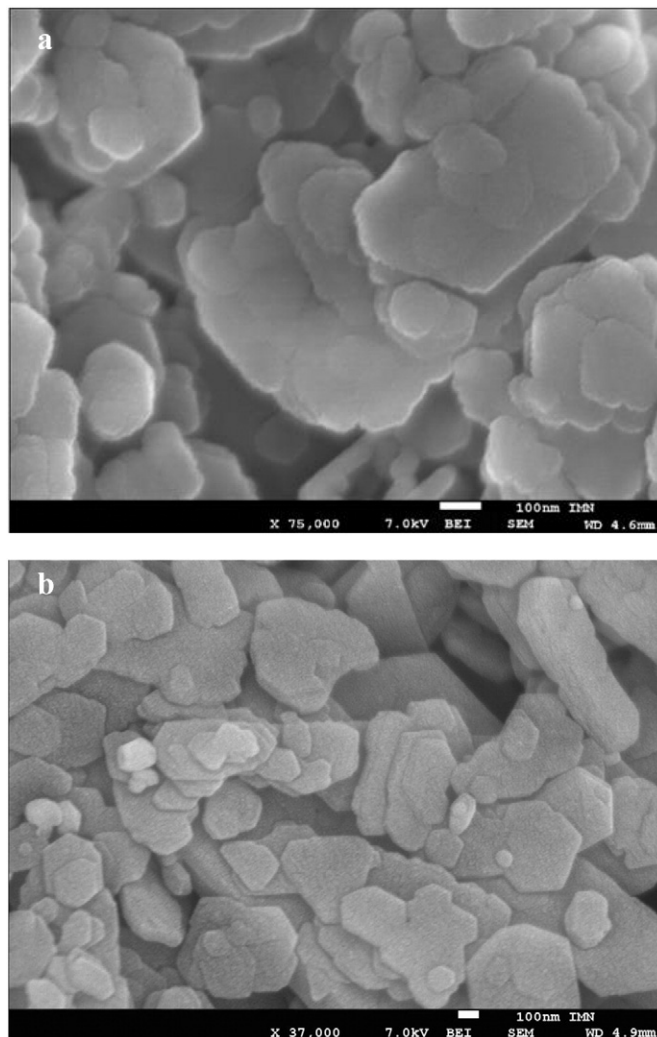


Fig. 1. SEM image of a) high defect, KGa-2 and b) low defect, KGa-1b Kaol.

The thickness of aggregated particles of the well-crystallized Kaol (KGa-1b) is higher than the poorly crystallized Kaol (KGa-2). The specific surface area, determined by nitrogen adsorption (BET method), was 9.95 and 18.6 ± 0.1 m²/g for KGa-1b and KGa-2, respectively suggesting that KGa-2 Kaol contains finer particles.

2.2. Zeta potential and size measurement

Electrophoretic mobility measurement was performed at 20 °C in a capillary cell with Malvern Nano Zetasizer apparatus and the Smoluchowski's equation was utilized to convert electrophoretic mobility to zeta potential. The measurements were carried out on agitated dispersions of Kaol particles (100 mg/l) dispersed in 10 mM NaCl solution. The pH of the dispersions was adjusted between 1 and 13 by adding either HCl, NaOH or Ca(OH)₂ solutions, equilibrated by agitation for 24 h in carbon dioxide free environment, and the pH was measured directly before introducing sample in to the capillary cell. Dynamic light scattering (DLS) measurements were performed at 20 °C and laser scattering at 173° to monitor the evolution of average particle size under different conditions (0 mmol/l, 22 mmol/l NaOH and 22 mmol/l Ca(OH)₂). For this purpose, the <1 μm fractions of Kaol particles were extracted by dispersing about 10 g of sample in 1 L ultra-pure water followed by centrifugation.

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