Multi-scale analysis on the effects of lime treatment on a kaolinite soil

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Abstract – **A relevant issue in the physical and hydro-mechanical behaviour of lime treated soil is the physico-chemical evolution of the system and its influence on the microstructural features of the treated soil. In the present study a multi-scale investigation on the short and long term effects of a lime treated kaolinite has been performed. Aim of the experimental work is the analysis of the link between the ongoing of the reactions induced by lime and the macroscopic evolution of soil properties. Some of the results obtained by means of oedometer tests on not treated and lime stabilised samples have been presented in the paper. The mechanical tests were complemented by investigations at microscopic level. The mineralogical changes induced by lime addition were monitored at increasing curing time by means of thermogravimetric analysis (TGA). The effects of lime addition on the clay particles arrangement were investigated by means of zeta potential and dynamic light scattering (DLS) measurements. Test results reveal a low reactivity of lime treated kaolinite to promote the development of pozzolanic reactions. The formation of hydrated phases was observed only starting from 28 days of curing proving that the improvement of soil properties is ruled by microstructure and not by bonding compounds resulting from pozzolanic reactions.**

I. INTRODUCTION

The study of the physical-chemical mechanisms controlling the behaviour of lime-stabilised soils is a relevant issue since the even more frequent use of this technique as an alternative and economical solution in geotechnical applications. The addition of lime to a clayey soils has a strong impact on the geotechnical properties of the soils as results of the chemical reactions which take place after the treatment. In the short-term, the exchange of surface cation by calcium leads to flocculation of clay particles. In addition, the high alkaline environment induced by lime promotes the dissolution of siliceous and aluminous compounds from clay mineral lattice with the precipitation of secondary phases as a result of the development of the time-dependent pozzolanic reactions [1], [2], [3], [4], [5]. In order to provide a better understanding of the link between the ongoing of the reactions induced by lime and the macroscopic evolution of soil properties, a multi-scale investigation on the short and long term effects of a lime treated kaolinite has been performed. In the present paper some oedometer test results on not treated and lime treated samples have been reported. The mechanical tests were complemented by investigations at microscopic level. The mineralogical changes induced by lime addition were monitored at increasing curing time by thermogravimetric analysis. The effects of lime addition on the clay particles arrangement were investigated by means of zeta potential and dynamic light scattering (DLS) measurements.

II. MATERIAL

The Speswhite kaolin, a highly refined kaolin from deposits in the South West of England, was the soil chosen for the present investigation. The specific gravity is 2.6 g/cm3, and the surface area determined by nitrogen adsorption (BET) is 14 m^2/g. The pH value of the soil is about 4.6. The liquid and plastic limits are respectively 70% and 32% (plastic index IP = 38%). The chemical composition of the soil is given in Table1.

Table 1. Chemical composition of Speswhite Kaolin

Constituent	Percentage (%)
SiO_2	53.80
Al_2O_3	43.75
Na_2O	0.92
CaO	0.02
K_2O	1.45
TiO_2	0.05

The bulk mineralogy of the soil (Figure 1) was determined by X-ray diffraction analysis performed on a randomly oriented sample using a Brucker AXS D8 Advance diffractometer with CuKα (λ=0.154 nm)

radiation and a step size of 0.021 degrees. X-ray diffraction patterns on the clay fraction (<2 μm) of air-dried and ethylene glycol saturated samples are shown in Figure 2. The soil is mainly formed by kaolinite clay minerals with a small amount of quartz and muscovite. No swelling clay phases were detected after the ethylene glycol treatment.
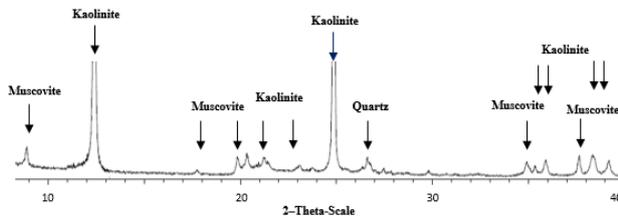

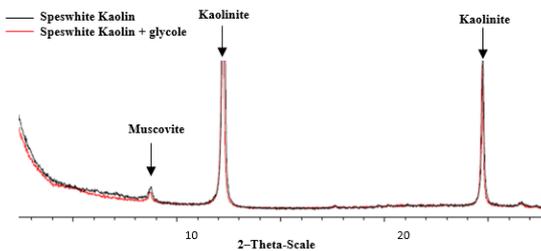*Fig. 1. X-ray diffraction pattern of Speswhite Kaolin.*

Fig. 2. X-ray diffraction pattern on clay fraction (<2μm).

III. EXPERIMENTAL PROCEDURES

A. pH measurements

Measurements of the Initial Consumption of Lime (ICL) were performed in order to assess the suitability of soil to lime treatment. The minimum amount of quicklime required for stabilization was determined by means of pH measurements according to the ASTM 4972, carried out on soil-lime-water mixtures (solid-liquid proportion of 1:1) prepared with different quicklime contents (namely 0.5%, 1%, 2%, 3%, 5% and 7% by weight of dry soil). Figure 3 shows the variation of pH values as a function of lime content obtained at increasing curing times (0 days (i.e. 24 hours), 14, 28, 60 days). The addition of 3% of quicklime was found sufficient to increase the pH to 12.4 (pH of a saturated lime solution) [6] and to maintain it high on the long term. Therefore, 3% of quicklime was selected as the percentage used for the samples preparation.
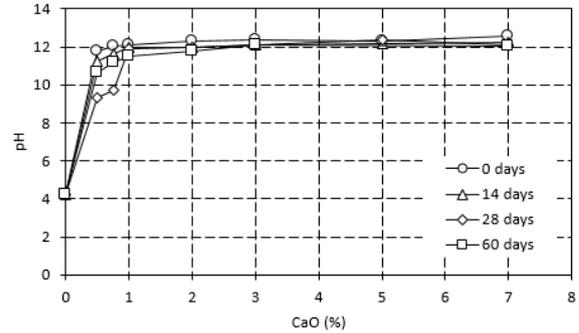

Fig. 3. pH measurements on lime treated kaolin..

B. Thermogravimetric analysis (TGA)

The mineralogical changes induced by lime addition were studied by TGA performed on not treated and lime treated samples at increasing curing times, namely 0, 3, 7, 14, 28, 60, 210 and 270 days. This technique measures the mass loss of the sample under a controlled atmosphere as a function of increasing temperature at constant heating rate. The change in mass is determined by the dehydration or decomposition of a mineralogical phase.

TGA was performed using a Netzsch STA 449F3 Jupiter apparatus. Samples were first dried by freeze-drying technique [7]. Approximatively 100 mg of finely grounded material was subsequently heated at a rate of $10°C$ min^{-1}, under argon atmosphere, from ambient temperature to 1000°C. The Netzsch Proteus software was used to process the results.

C. Zeta potential and dynamic light scattering (DLS) measurements

The surface charge properties of kaolinite particles and the evolution of the average dimension of particles agglomerates over the time were investigated by means of zeta potential and dynamic light scattering (DLS) measurements. The zeta potential was evaluated from the electrophoretic mobility using the Smoluchowski formula [8]. The electrophoretic mobility was measured in a capillary cell using a Malvern Nano Zetasizer apparatus thermostated at constant temperature T = 25 °C. The measurements were carried out on kaolinite suspensions (100mg/l) as a function of pH. The pH of the dispersion was adjusted to values between 2 and 12.4 by adding either solutions of HCl, KOH or $Ca(OH)_2$. The Malvern Nano Zetasizer apparatus was also used to perform dynamic light scattering measurements. This technique allows monitoring the evolution of particle aggregation process over the time. Measurements were made at T = 25°C with a scattering angle of 173° on kaolinite suspension (100mg/l) in de-ionized water and in presence of KOH and $Ca(OH)_2$ solutions at pH 12.4.

D. Oedometer tests

Oedometer tests were performed on not treated and lime stabilised samples cured for increasing time intervals. Hand remoulded samples were prepared by mixing the soil with distilled water to a slurry at about its liquid limit. The same procedure was used for lime treated samples prepared starting from slurry condition at an initial water content corresponding to the liquid limit of the stabilised samples (w_L=101%) and then placed in the oedometer mould without compaction. The treated samples were sealed in plastic bags and cured at increasing curing times of 0, 14 and 28 days. The tests were performed in standard oedometer cells, where vertical stress was conventionally applied in successive steps ($\Delta\sigma v/\sigma v$=1).

IV. RESULTS

The zeta potential of kaolinite slurries as a function of pH is shown in Figure 4.

The magnitude of this parameter is considered as a measure of the particle repulsions [9] and therefore depends on the surface charge properties of kaolinite particles. In the absence of calcium, the magnitude of zeta potential increases with increasing pH and decreases with decreasing pH as a result of the deprotonation and protonation reactions on the kaolinite edge and octahedral basal surfaces. In the presence of calcium (i.e. with increasing the concentration of calcium hydroxide), the magnitude of zeta potential decreases with increasing pH. The observed behaviour can be linked to calcium adsoption at the surfaces of the negatively charged particles which leads to a reduction of the net negative charge on particle surfaces.
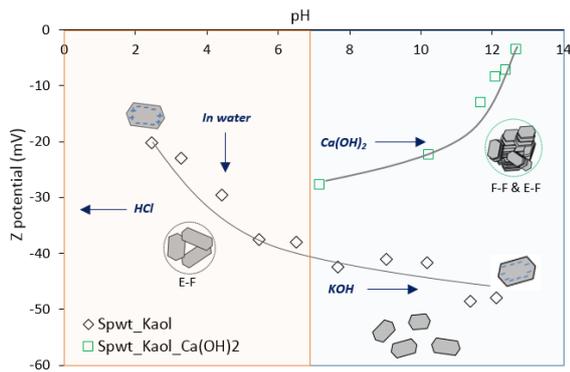

Fig. 4. Zeta potential measurements.

In order to gain further insight into the kaolinite particles interactions and flocculation behaviour, the evolution of the particles aggregation process with time was monitored under different testing conditions by means of dynamic light scattering (DLS) measurements. At natural pH of kaolinite suspension in deionized water (pH = 4.6) (Figure 5), the measured average floc size increases with time as a consequence of positively charged edge-to-negatively charged basal face mode of particle aggregation [10]. When the pH increases to 12.4 (by adding KOH), the size decreases and remains unchanged over time suggesting a dispersed state of kaolinite particles. The high negative surface charge, as indicated by higher zeta potential (Figure 4), and a consequent increase of the electrostatic repulsive forces prevents particle collisions and aggregation. In the presence of calcium (Ca(OH)$_2$) and high pH (pH = 12.4) the size of kaolinite flocs is higher than the case of deionized water and KOH solution. The floc size increases with time revealing the formation of a strongly flocculated structure. The calcium ions counteract the negative surface charge and repulsive forces promoting particles aggregation.
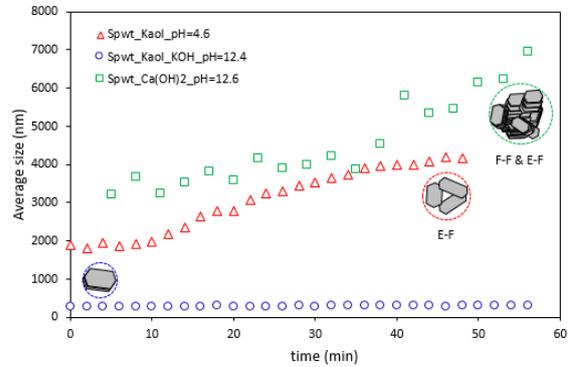

Fig. 5. DLS measurements of kaolinite suspension under different chemical environments.

The evolution of the system after the addition of lime has been also investigated from a mineralogical point of view by means of thermogravimetric analysis. **Errore. L'origine riferimento non è stata trovata.**Figure 6 shows the mass loss of the sample as a function of curing time in the temperature range of 390°C-460°C, which is characteristic of portlandite and in the range of 130°C - 350°C typical of the dehydration of Ca-hydrates. A delay between the portlandite consumption and the formation of new hydrated phases reveals the slow reactivity of kaolinite to promote the development of pozzolanic reaction. The formation of secondary phases appeared to form only after a long curing time (> 28days).
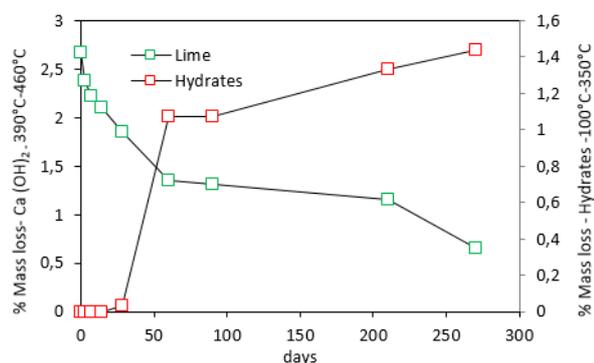
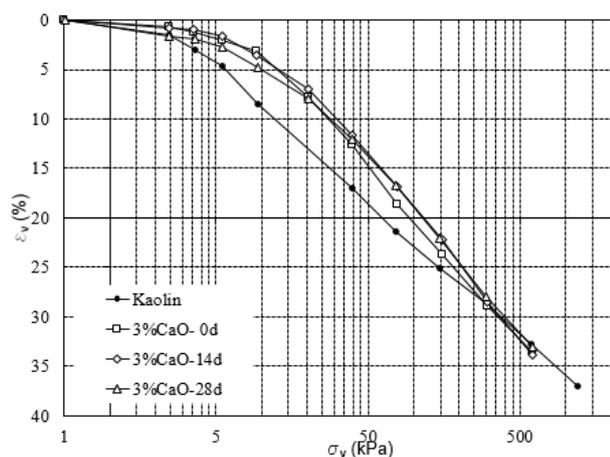*Fig. 6. Mineralogical evolution of lime stabilised samples.*

Fig. 7. Oedometer test results on not treated and lime stabilised samples at increasing curing times.

The physico-chemical evolution of the system induced by the addition of lime has been linked to the macroscopic evolution of soil properties. One dimensional compressibility curves of not treated and lime treated samples at 0, 14 and 28 days of curing times are shown in Figure 7. The addition of lime induces a modification of the observed behaviour with a reduction of the volume strains and an increase of the yield stress of the treated samples. No relevant changes were detected at increasing curing times. This result seems to be consistent with the mineralogical and microstructural evolution of the system. The mechanical improvement of the treated soil is only affected by the microstructural re-organisation of clay particles since the lack of formation of new phases in the short term, as highlighted by the experimental results.

V.   CONCLUSION

In the present paper a multi-scale investigation on the short and long term effects induced by lime on a kaolinitic soil has been proposed in order to investigate the link between the ongoing of the reactions and the macroscopic evolution of the soil properties. Test results show the formation of an aggregated particle structure in lime treated kaolinite suspensions as a result of the flocculation phenomena induced by ionic exchange reactions. Slight changes in the mechanical properties (reduction of the compressibility with an increase of the yielding stress) were observed after lime addition at increasing curing time, due to the no significant pozzolanic activity taking place until 28 day of curing. The low reactivity of kaolinite soil is reflected in a delay of the portlandite consumption and the formation of new phases as revealed by the mineralogical investigations. The formation of hydrate phases was observed only after 28 days of curing proving that the improvement of soil properties is ruled by microstructure and not by bonding compounds resulting from pozzolanic reactions.

REFERENCES

[1]   F.G. Bell, "Lime stabilization of clay minerals and soils", Engineering Geology, 1996, 42, 223-237.

[2]   S. Diamond, E. B. Kinter, "Mechanisms of soil lime stabilization: an interpretative review," Highway Research Records, 1965, 92, pp. 83-96.

[3]   C. D. F. Rogers, S. Glendinning "Modification of clay soils using lime", In Lime Stabilisation,1996, C. D. F. Rogers, S. Glendinning and N. Dixon (eds.), 99-112, Thomas Telford Academic Press, New York, USA, 2009.

[4]   G. Russo, G. Modoni, "Fabric changes induced by lime addition on compacted alluvial soil", 2013, Géotechnique Letters, Vol. 3, Issue 2.

[5]   K. Lemaire, D. Deneele, S. Bonnet, M. Legret, " Effects of lime and cement treatment on the physicochemical, microstructural and mechanical characteristics of a plastic silt, Eng. Geol., 2013, 166, 255-261.

[6]   J. L. Eades, R. E. Grim, " A quick test to determine lime requirements for lime stabilisation", Highway Research Board Record, TRID, 1966, 139, pp. 61-72

[7]   P. Delage, F. M. Pellerin, "Influence de la lyophilisation sur la structure d'une argile sensible du Québec". Clay minerals, 1984, 19, 151-160.

[8]   R. J. Hunter, "Zeta Potential in Colloid Science", Academic Press, New York, 1981.

[9]   H. Van Olphen, " Clay colloid Chemistry: For Clay Technologists, Geologists and Soil Scientists", John Wiley, 1977.
      E. Tombacz, M. Szekeres, "Surface charge heterogeneity of kaolinite in aqueous suspension in comparison with montmorillonite suspensions". Appl. Clay Sci., 2006, 34, 105-124.