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# ELASTIC MODULUS OF SOILS TREATED WITH LIGNOSULFONATE

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## ABSTRACT

This paper presents the variation of modulus, in terms of  $E/q_u$ , with strain for clay soils stabilised by a non-traditional admixture, lignosulfonate. The  $E/q_u$  has been computed from the stress-strain curves obtained from unconfined compressive strength tests. Lignosulfonate (LS) treated soil shows an increase in the value of  $E/q_u$  with the addition of LS, especially for soil containing the reactive clay mineral, montmorillonite. The observed variation of  $E/q_u$  with strain was compared with cement treated soils, and it was found that soils treated with more than 1.0% cement exhibited a decreasing  $E/q_u$ . Moreover, micro-chemical changes in the soils due to the addition of admixture (e.g. LS & Cement) were captured using electrical conductivity tests. Electrical Conductivity (EC) is found to be increased with LS and decreased over time. This may be due to the formation of clay mineral-lignosulfonate amorphous compounds, which are stable when comes in contact with water. However, cement treated soils showed an increase in EC with curing time, unlike LS that confirmed the existence of unstable compounds (i.e. dissolvable CSH and CASH) resulting in a decrease of  $E/q_u$ .

*Keywords:* mineralogy, cement, lignosulfonate, brittle, ductile, electrical conductivity

## 1 INTRODUCTION

Chemical admixtures such as cement and lime are widely used as additives to stabilise clay soils. These admixtures generally alter the mineralogical structure of clay and improve its inherent properties such as strength and stiffness. However, all chemical admixtures are not frequently useable because they alter the pH of soil, which results in a corrosive environment (Sherwood, 1993), and they often limit the scope of re-vegetation and cause a threat to ground water pollution. Moreover, traditionally treated soils have been shown to be brittle behaviour, which is often undesirable for structures, especially those under cyclic loads such as rail embankments and aircraft runways. To overcome these difficulties, researchers are now looking for new soil additives, which can overcome most of the adverse effects of traditional admixtures, apart from maintaining sufficient ductile properties of the soils.

In this context lignosulfonate has shown some promising prospects as a soil admixture, especially for clay soils. Puppala and Hanchanloet (1999) and Tingle and Santori (2003) carried out laboratory investigations on cohesive soils with lignosulfonate and showed that lignosulfonate accompanied with a small amount sulphuric acid, greatly improved the shear strength and modulus resilient of the soil. Tingle and Santori (2003) found that lignosulfonates are capable of improving the strength of clayey soils with low plasticity. A number of researches (e.g. Chemstab 2003, Tingle and Santori 2003) were performed to investigate whether such lignin-based products could be used in low volumes for road construction to improve the strength of sub-grade and control the emission of surface dust. Recently, Indraratna et al. (2008) conducted research on the internal erosion of dispersive soils treated with lignosulfonate and highlighted that this treatment improved the resistance of treated soil to erosion, just like traditional admixtures such as cement. However, the improvement of soil stiffness due to the treatment of lignosulfonate still needs to be investigated, so in this study an attempt has been made to investigate the improvement of modulus (i.e.  $E/q_u$ ) of soils due to treatment by lignosulfonate. Furthermore, micro-chemical analysis was also performed on treated soils to understand the mechanism of stabilisation.

## 2 STABILISATION MECHANISM OF LIGNOSULFONATE AND CEMENT

Lignosulfonate is a lignin based polymeric compound (e.g. [Mollah et al. 1995](#); [Lemes et al. 2005](#)) derived as a waste product from the wood/paper processing industry. It consists of both hydrophilic groups including sulphonate, phenolic hydroxyl, and alcoholic hydroxyl, and hydrophobic groups including the carbon chain (e.g. [Chen, 2004](#)). The molecular weight of lignosulfonate varies from 4600 to 398000 g/mol ([Fredheim et al. 2002](#)) and these are soluble in water over the entire range of pH. The molecular weight and structure of lignosulfonate depends on sources of extraction such as soft wood (i.e., Pine, Fir), hard wood (i.e., Eucalyptus, Beech, Aspen), grass, and paper mill. The main advantages of lignosulfonate over traditional stabilisers are non-toxicity and non-corrosiveness. Moreover, it is not classified as bio-degradable hydrocarbon like bacteria, yeast, or fungi. The stabilisation mechanism of lignosulfonate is based on the electro-static reaction process. When lignosulfonate is mixed with water it forms positively charged ions through hydrolysis reaction. This positively charged LS reacts with negatively charged clay minerals that reduces the double layer thickness of the clay particles to form stable particle clusters or aggregates ([Vinod et al. 2010](#)). The electro-static reaction depends on the surface charge of clay minerals and chargeability of the admixtures. So mineralogy of soils is a very important aspect for stabilisation by lignosulfonate.

The stabilisation mechanism of a traditional admixture such as cement is based on the formation of primary and secondary cementitious compounds in stabilised soils. The primary cementitious compounds are formed by a hydration reaction comprised of hydrated calcium silicates, calcium aluminates, and hydrated lime ([Bergado et al. 1996](#); [Schaefer et al. 1997](#)). The presence of platy and formation of reticular cementitious products (i.e., CSH and CASH) in the stabilised soil is mainly responsible for the change of physico-chemical properties and development of long term strength ([Umesha et al. 2009](#)). Compounds such as silica and alumina that participated in the secondary reaction are contributed to by the clay minerals. So the mineralogy of soils has a significant influence on reaction and outcome of chemically treated soils. Previous studies showed that montmorillonite is active and kaolinite is comparatively an inert clay mineral. The reaction of montmorillonite with cement is faster than other clay minerals and has a profound influence on the hardening of treated soils. Kaolinite, illite ([Porbaha et al. 2000](#)), and other well organised (well crystallised ) clay minerals have little effect in the hydration reaction with cement ([Stavridakis 2005](#)) and thus these clay minerals require a large amount of cement for this reaction to achieve satisfactory strength and durability ([Bell 1976](#); [Croft 1967](#)). It has been reported that kaolinite is rapidly exhausted by the pozzolanic reaction for 10% of cement treatment ([Eades and Grim 1960](#); [Kamruzzaman et al. 2006](#)). This is because with a higher content of cement, the silicates and aluminates from the kaolinite go into the solution where they react with the adsorbed  $Ca^{2+}$  ions on the surface of clay mineral to form CSH and CASH, which in turn induces cementation of the flocculated clay particles to form clay-cement clusters ([Rao and Rajasekaran 1996](#); [Kamruzzaman et al. 2006](#)). Thus both the dissolution of the kaolinite and the flocculation process result in more clay-cement clusters interspersed by large voids. While considering the stabilisation mechanism of traditional admixtures, it can be said that the clay mineralogy of soil has significant influence in improving the strength and stiffness of treated soils, including the brittle-ductile transition. This may be due to changes in its physical and strength properties with electrical properties such as electrical conductivity and pH (e.g. [Boardman et al. 2001](#); [Chen et al. 2006](#)). Therefore, an attempt was made in this study to investigate the modulus behaviour of chemically treated soil based on soil mineralogy and the electrical conductivity of pore fluids.

## 3 MATERIALS AND METHODS

Two soil samples, a kaolinite and a clay mixture consisting of 95% kaolinite and 5% montmorillonite, were used in this study. The index properties of the soils are presented in [Table 1](#). The compaction tests were performed on the samples using mini compaction apparatus ([Sridharan and Sivapullaiah 2005](#)) and the values are presented in [Table 1](#). It shows that the maximum dry density decreased and the optimum moisture content increased with the content of montmorillonite. This was due to the presence of a hydrous clay mineral i.e., montmorillonite, which has a large surface area and a high capacity to absorb moisture compared to kaolin ([Sridharan and Jayadeva 1982](#)). The presence of montmorillonite in soils increases the water retention capabilities, which results in a higher value of optimum moisture content and lowers the value of maximum dry density (e.g., [de Magistris et al. 1998](#)).

Table 1. The index properties of soils used in this study

Soil sample	$G_s$	LL (%)	PL (%)	Compaction properties	
				MDD ( $\text{kg}/\text{m}^3$ )	OMC (%)
Kaolin clay	2.62	54	31	1498	26.7
Clay mixture (95 % Kaolin + 5 % Montmorillonite)	2.64	56	26	1480	27.8

$G_s$ -specific gravity; LL-liquid limit; PL-plastic limit; MDD-maximum dry density; and OPC-optimum moisture content.

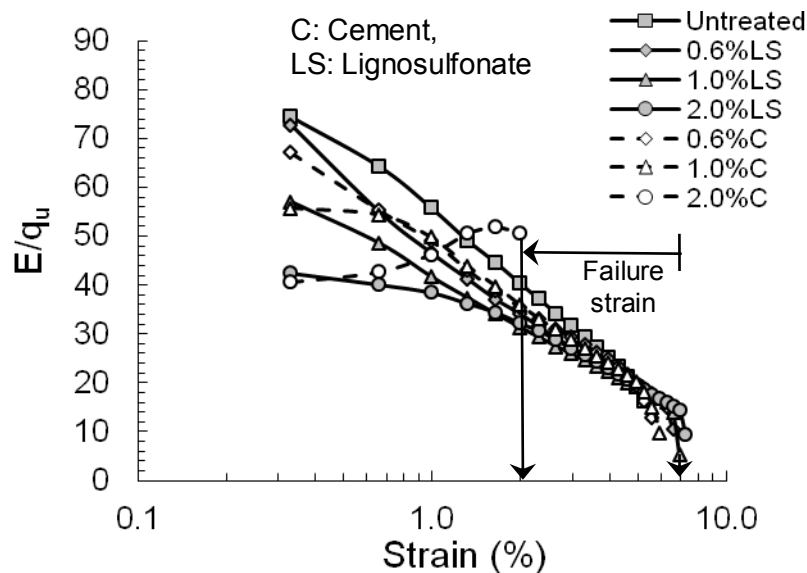


Figure 1. Variation of modulus ( $E/q_u$ ) for kaolin clay treated with lignosulfonate and cement

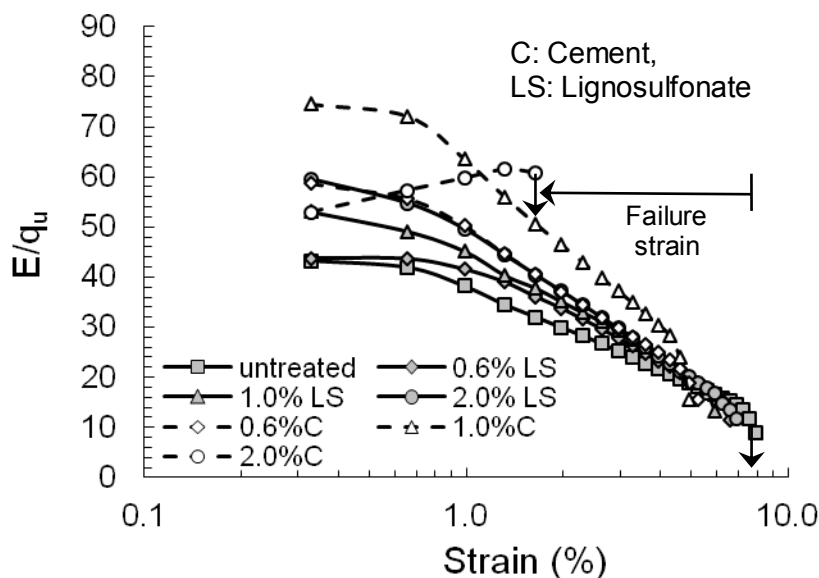


Figure 2. Variation of modulus ( $E/q_u$ ) for clay mixture treated with lignosulfonate and cement

#### 4 TEST RESULTS AND ANALYSIS

The modulus,  $E/q_u$ , of soil treated with lignosulfonate (LS) and cement was studied using standard unconfined compressive strength tests. The samples for the tests were prepared at maximum dry density (i.e. optimum moisture content) with different amounts of chemical admixtures (0.6%, 1.0%, and 2.0%). The treated and untreated samples of soil were cured in a humidity controlled room at a temperature of 20°C for 7 days. The variation of modulus for treated and untreated soils is presented by plotting the results in the form of  $E/q_u$  versus  $\log \epsilon$ , where,  $E$ ,  $q_u$ , and  $\epsilon$  are secant Young's

modulus, ultimate strength, and axial strain, respectively. The secant modulus was calculated over the entire range of the stress-strain curve (i.e., up to failure stress of sample). The variation of  $E/q_u$  with respect to axial strain for treated and untreated soils is presented in Figs. 1 & 2, where it is found that the behaviour of the modulus was clearly non-linear.

For kaolin clay, the initial value of  $E/q_u$  decreased with an increase of lignosulfonate (LS), which is similar to the cement treated soil (Fig. 1). The failure strain for untreated kaolin was close to 6.0%. After treating the kaolin clay with different percentages of LS, the failure strain was almost the same as untreated soil. On the other hand, the failure strain of soil treated with 0.6% and 1.0% cement showed behaviour that was similar to LS treated soil. However, a further increase of cement i.e. treatment with cement at 2.0%, reduced the failure strain to 2%, which was much lower than the untreated and other treated soils. Thus, kaolin clay becomes very brittle when treated with 2.0% cement, however the same percentage of LS treatment did not reduce the failure strain of soil compared to cement.

For the case of Clay mixtures, the initial value of  $E/q_u$  was found to increase with the addition of LS. The clay mixture treated with cement showed a higher value of  $E/q_u$  compared LS (Fig. 2). However, the soil treated with 2.0% cement had a lower value compared to the soil treated with 1.0% cement. On the other hand the failure strain for untreated soil was found as 8.0% but it decreased to nearly 6% due to treatment with LS. For the case of cement treatment, the failure strain of 0.6% and 1.0% was almost 5%, which is very close to LS treated soil. However, when clay mixture was treated with 2.0% of cement, the failure strain was found as 1.7%, which is much lower than LS treated soils. Based on this observed behaviour, it can be said that the mineralogy of soil, type of additive, and content of additive have a significant influence on the modulus behaviour of treated soils. Further chemical analysis was performed to explain the observed behaviour of treated soils.

## 5 CHEMICAL ANALYSIS OF TREATED AND UNTREATED SOILS

The Electrical Conductivity (EC) for treated and untreated soils was measured using an electrical conductivity meter in 1:5 (soil: water) suspension to find out the ionic concentration of pore fluid in soil matrix. The soil-water suspension was prepared by mixing 10 g air-dried soil (<2 mm) with 50 ml de-ionised water. The soil-water mixture was stirred with a magnetic stirrer for 10 minutes to completely dissolve the soluble salts in water. Then the EC of soil suspension was measured with an electrical conductivity meter. The electrical conductivity test results for treated soils are presented in Figs. 3-5. It is observed that the EC of LS treated soil increased with an increase of additive (Fig. 3). With the increase of curing time, the EC for LS treated soils decreased over the curing of 7 days but then remained almost constant over a wide range of curing time (Fig. 4 & 5). The increasing EC of pore fluid of treated soil indicated a decrease of dielectric constant (Kaya and Fang 1997) that caused a contraction of the double layer thickness of soil particles to contract and also to flocculate (Bowders *et al.*, 1985). The decrease of EC with curing time indicates the formation of clay mineral-lignosulfonate amorphous compounds, which are stable in contact with water and thus form stable soil aggregates.

On the other hand, when the additive was increased, the EC for soils treated with cement was similar to LS (Fig. 2), although the variation of these soils with curing time was different compared to LS. The EC trend for soils treated with 1.0% cement was similar to LS, but the behaviour of soils treated with 2.0% cement was different from lignosulfonate treated soils (Figs 4 & 5). The EC for soils treated with 2.0% cement increased exponentially during the curing period of 7 days, but decreased slightly over the curing of 28 days. The decreasing trend of EC with curing time indicated the formation of stable compounds in treated soils. On the other hand, the increase of EC with 2.0% cement indicated the existence of excess ions in the pore fluid. The sources of excess ions may be unstable CSH and CASH (still dissolvable in water), and a reorientation of the clay minerals' structure (i.e., exploitation of lattice). This structural reorientation (e.g. exploitation) of soil mineral with curing time due to treatment with cement was explained by Kamruzzaman *et al.* (2006). The modulus behaviour of cement treated soils can be related to variations in their electrical conductivity. When soil is treated with a small amount of cement the formation of cementitious compounds completely reacts with the surface of the clay to bind the soil particles together and forms stable compounds (i.e. not dissolved in contact of water). In this case, cement works as a binding and a filling material. It results in a decrease of EC with curing time and results in a higher ultimate strength while maintaining its ductile behaviour. However, when soils are treated with higher amounts of additive, it forms higher amount of

cementitious compounds in soils that may not get enough reactive clay mineral for a reaction. The excessive amount of cementitious compounds produces reticulation (opening structure) resulting in the reduction of modulus and failure strain and thus exhibits brittle behaviour.

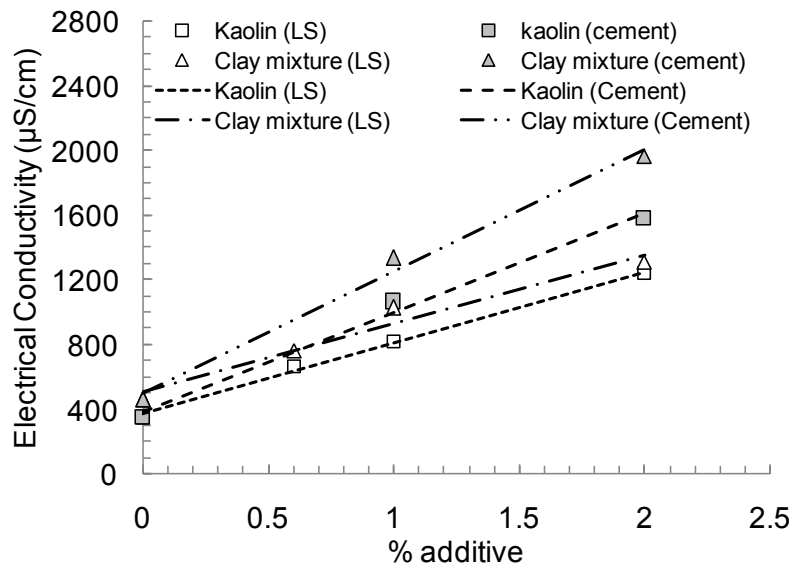


Figure 3. Variation of electrical conductivity for treated soils

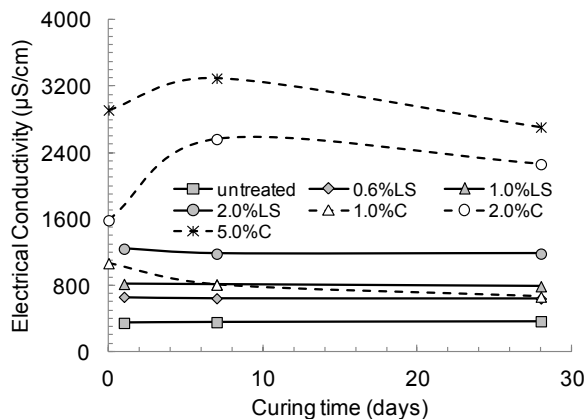


Figure 4. Variation of electrical conductivity of treated kaolin with curing time

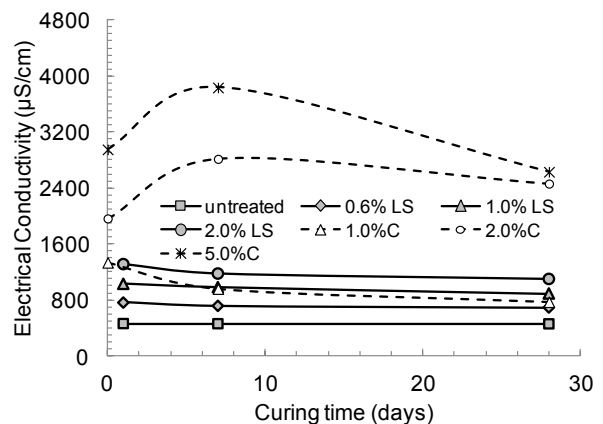


Figure 5. Variation of electrical conductivity of treated clay mixture with curing time

## 6 CONCLUSION

This paper highlights the potential of lignosulfonate as a soil admixture for stabilising clay soils. It was found that the modulus  $E/q_u$ , of soils treated with LS increased with the admixture without reducing the failure strain, especially for soils containing the reactive clay mineral, montmorillonite. The modulus behaviour was compared with cement treated soils, where it was found that soils treated with cement ( $\leq 1\%$ ) showed a variation in the modulus that was similar to LS. However, soils treated with more than 1.0% cement showed a high reduction in the modulus as well as failure strain, unlike LS. The electrical conductivity tests showed that the Electrical Conductivity (EC) of treated soil increased with LS and decreased over time. This was mainly due to the reaction and formation of stable compounds that improved the modulus while maintaining its ductile behaviour. However, the EC of soils treated with cement ( $> 1.0\%$ ) showed an increasing trend with curing time, unlike LS. This confirmed the existence of unstable compounds that reduced the modulus and failure strain and were thus exhibiting high brittle behaviour compared to lignosulfonate treated soils.

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