SOIL STABILIZATION USING LIGNOSULFONATE

Jayan S. Vinod[1], Buddhima Indraratna[2] and Ana Heitor[3]
[1]Senior Lecturer,[2]Distingushed Professor and Research Director, [3]Lecturer
Centre for Geomechanics and Railway Engineering; ARC Centre of Excellence for Geotechnical Science and Engineering, University of Wollongong, Wollongong, Australia
vinod@uow.edu.au; indra@uow.edu.au;aheitor@uow.edu.au

ABSTRACT

Chemical stabilisation of soil using commercial admixtures is a common technique often adopted by industry to improve the behaviour of erodible and unstable deposits such as those forming the subgrade for roads and rail infrastructure. Although traditional alkaline admixtures such as cement, gypsum, lime have been effective in strengthening and stiffening the natural formations of transport and other civil infrastructure, the consequential impact on the soil and groundwater chemistry has been an environmental concern for many years. While being cost-effective, these alkaline and sometimes corrosive admixtures have directly contributed to substantial rise in the soil and groundwater alkalinity (pH about 8-9) apart from the significant reduction in soil porosity (void space) thus adversely affecting the growth and development of certain native vegetation and sub-surface fauna. In contrast, the use of non-toxic lignosulfonates (LS) with much smaller quantities has been reported to achieve similar results without harming the environment.

Lignosulfonate is a soluble dark brown liquid, and it is a by-product of the timber and paper industry. The use of lignosulfonate as a soil stabilizer has significant advantages in relation to traditional admixtures with respect to soil and groundwater environment quality. This is because lignosulfonate is readily diluted in water and causes no pH change in the soil upon treatment. Furthermore, it is also non-corrosive to metals and non-flammable, and classified as non-hazardous. An additional benefit is associated with the reduction of brittle behaviour during shear loading that is well known for alkaline mixtures. This paper showcases a number of applications of LS in controlling erosion and swelling of expansive soil for which the use of lignosulfonate has yielded comparable and in cases superior performance to that of traditional admixtures.

1 INTRODUCTION

In many parts of the world, the incidence of problematic soft soils including erodible, dispersive and expansive soils causes damage to infrastructure that severely impacts its operation. Chemical stabilisation using traditional admixtures such as cement, lime, fly ash is generally used for stabilising these soils. Typically, these chemical stabilisers generally alter the mineralogical properties of the soil resulting in highly stable soil substances with improved engineering properties such as strength and stiffness. A number of studies have been reported to investigate the applicability of traditional stabilisers on problematic soils such as soft clay and erodible soils (e.g. Balasubramaniam *et al.*, 1989; Indraratna *et al.*, 1995; Rajasekaran *et al.*, 1997; Uddin *et al.*, 1997; Chew *et al.*, 2004).

While, the use of these traditional admixtures (i.e. cement, lime, fly ash) has been relatively well established in practice, they can cause various threats to the environment due to inevitable increase in soil and groundwater alkalinity. For instance, Rollings et al (1999) highlighted that traditionally stabilised soil has a pH of 9, which can often affects the longevity of concrete reinforcement and steel frame structures (e.g. Biggs and Mahony, 2004; Perry, 1977). In addition, other chemical aspects such as electrical conductivity and cation exchange capacity of the soil decreases with the increasing amount of admixtures and curing time (e.g. Chen *et al.* 2009; Boardman *et al.,* 2001). This may influence the water retention capacity as well as nutrients carrying capacity of soil resulting in lower fertility (e.g. Lund *et al*., 1999; Kitchen and Sudduth, 1996; Jaynes *et al*., 1995). Therefore, the excess use of traditional admixtures in soil stabilisation could be harmful for the yielding capacity of soils (e.g. Nalbantoglu and Tuncer, 2001). Furthermore, the traditionally treated soil exhibits excessive brittle performance (e.g. Sariosseiri and Muhunthan, 2009) that affects the stability of structures, especially during cyclic and impact loading conditions such as high speed rail and aircraft runways. In this context, an alternative soil stabiliser, which could provide sustainable soil improvement without harming the environment, would be preferable.

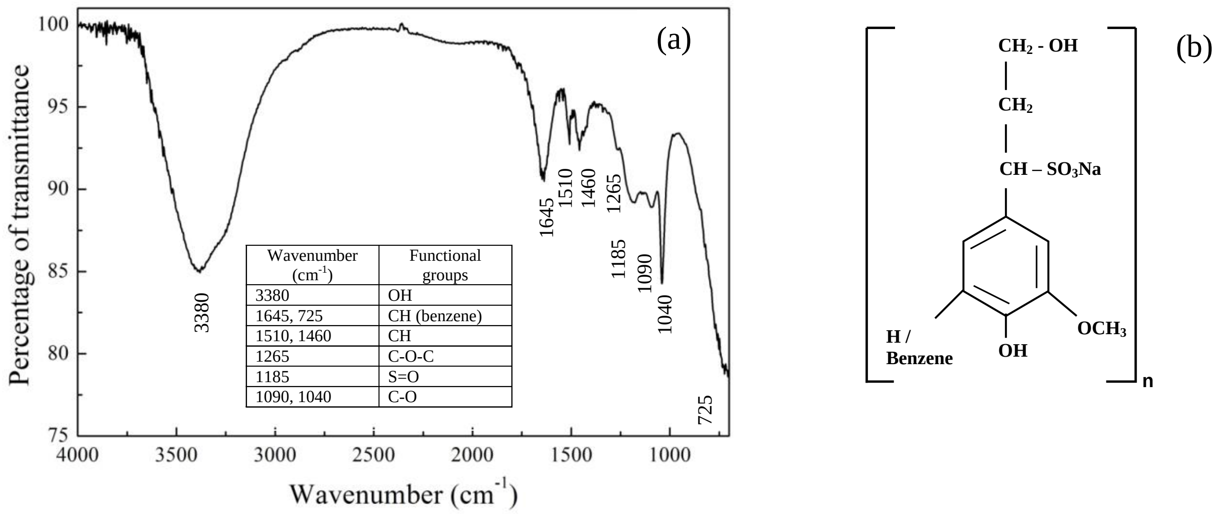
Recently, a lignin based chemical, i.e. lignosulfonate has shown promising results in stabilising the problematic soils (Puppala and Hanchanloet, 1999; Pengelly *et al.*, 1997; Tingle and Santori, 2003; Indraratna *et al*., 2008; Vinod et al.2010). In addition, a number of research studies have been conducted on low volume road construction to investigate the performance of lignosulfonate for improving the strength behaviour of subgrade and also for effective control of dust emissions (e.g. Chemstab, 2003; Tingle and Santori, 2003; Lohnes and Coree, 2002). Sherard *et al.* (1976) studied the erosion characteristics of soil by directing eroding fluid through a 1-mm crack (hole), i.e. standard pinhole test. Wan and Fell (2004) performed erosion tests by applying a hydraulic gradient across a 6 mm internal crack to study the erosion characteristics of unsaturated soil in cracks of embankment dams. Pengelly *et al.* (1997) carried out studies on expansive soils by injecting a chemical mixture consisting of ammonium lignosulfonate and potassium chloride and observed a significant reduction in swelling potential.

Research carried out at University of Wollongong (UOW) over the years also shows that LS improves the erosion resistance of erodible soils and has a comparable performance in relation to traditional admixtures such as lime and cement (Indraratna et al., 2008, Indraratna et al., 2013). More recently, lignosulfanate has also proven to be effective in controlling the swelling potential of expansive soils (Alazigha et al., 2016). In this paper the key research outcomes on the use of lignosulfonate and associated effectiveness in stabilising erodible and expansive soils is presented. A mathematical model that captures the erosion behaviour of chemically treated erodible soil is also presented.

2 LIGNOSULFONATE ADMIXTURE

Lignosulfonate (LS) is a lignin based polymeric stabiliser is waste by-product derived from the wood/paper industry (e.g. Mollah et al., 1995; Lemes et al., 2005). It consists of both hydrophilic groups including sulphonate, phenylic hydroxyl, and alcoholic hydroxyl and hydrophobic groups including carbon chain (e.g. Chen, 2004). The molecular weight of lignosulfonate varies from 4600 to 398000 g/mol (Fredheim et al., 2002) and it is soluble in water over the entire range of pH. The molecular weight and structure of lignosulfonate depends mainly on the type of sources of its extraction, for instance soft wood (i.e., Pine, Fir), hard wood (i.e., Eucalyptus, Beech, Aspen), grass, and paper mill.. In addition, the molecular weight increases with the decrease of the relative amount of sulphonic acid groups. The molecular weight influences the bonding properties of lignosulfonate.

The functional groups of LS can be studied using Fourier Transform Infrared Spectroscopy (FTIR). Fig 1 (a) shows the FTIR results of a LS sample that was scanned for wave number ranging from 4000 to 800 cm^{-1} at resolution of 16 cm^{-1} and scan interval of 4 cm^{-1}. It is evident from Fig. 1(a) that the lignosulfonate contains the functional groups such as OH (3380 cm^{-1}), benzene ring (1645, 1510, 725 cm^{-1}), C–H stretching group (1460 cm^{-1}), C–O bond primary alcoholic group (1090 cm^{-1}), C–O bond secondary alcoholic group (1040 cm^{-1}), C–O–C stretching –OCH3 group (1265 cm^{-1}) and S=O stretching sulfonate group (1185 cm^{-1}). Based on the observed functional groups and chemical composition, the structure of lignosulfonate is developed and is presented in Fig. 1b. More details on the structure of LS can be found elsewhere (e.g. Vinod et al.2010 and Algahzia et al., 2016)



Wavenumber (cm^{-1})	Functional groups
3380	OH
1645, 725	CH (benzene)
1510, 1460	CH
1265	C-O-C
1185	S=O
1090, 1040	C-O

Figure.1: (a) FTIR analysis of lignosulfonate, (b) structure of lignosulfonate (Vinod et al., 2010)

3. ERODIBLE SOIL STABILISATION USING LIGNOSULFONATE

At UOW, studies on internal crack erosion were carried out using the Process Simulation Apparatus for Internal Crack Erosion (PSAICE). Detailed explanation of testing apparatus and testing procedure can be found elsewhere (e.g. Indraratna *et al*. 2008). The erosion behaviour of treated and untreated soils have been examined in terms of erosion rate and hydraulic shear stress to calculate the erosion parameters, namely, the critical shear stress and the coefficient of soil erosion. The soil used in this study was highly erodible soil from the Wombeyan caves (NSW) classified as a silty sand (LL=22.5%, non-plastic) and D1 erodibility class (AS 1289.3.8.3, 1997) The critical shear stress, τ_c is defined as the minimum hydraulic shear stress required to initiate erosion. Fig. 2shows the variation of the erosion rate with the hydraulic shear stress. The critical shear stress was calculated by extrapolating the straight line to the X-axis, and the slope of the linear line represents the coefficient of soil erosion. It is evident that the erosion rate and hydraulic shear stress follow a linear relationship, and the slope represents the coefficient of soil erosion. It can be observed that the critical shear stress increases and the coefficient of soil erosion decreases with the increase of lignosulfonate (Fig 2a), i.e. the critical shear stress increases from 0.8 Pa to 35 Pa with the addition of 0.6% LS. Conversely, the critical shear stress increases from 0.8 Pa to 43.4 Pa with addition of 3% cement. Fig 2 results indicate that not only the addition of LS results in comparable performance to that of traditional admixtures (cement), the amount of LS required to achieve high critical shear stresses is much smaller (LS 0.6% and cement 3%).

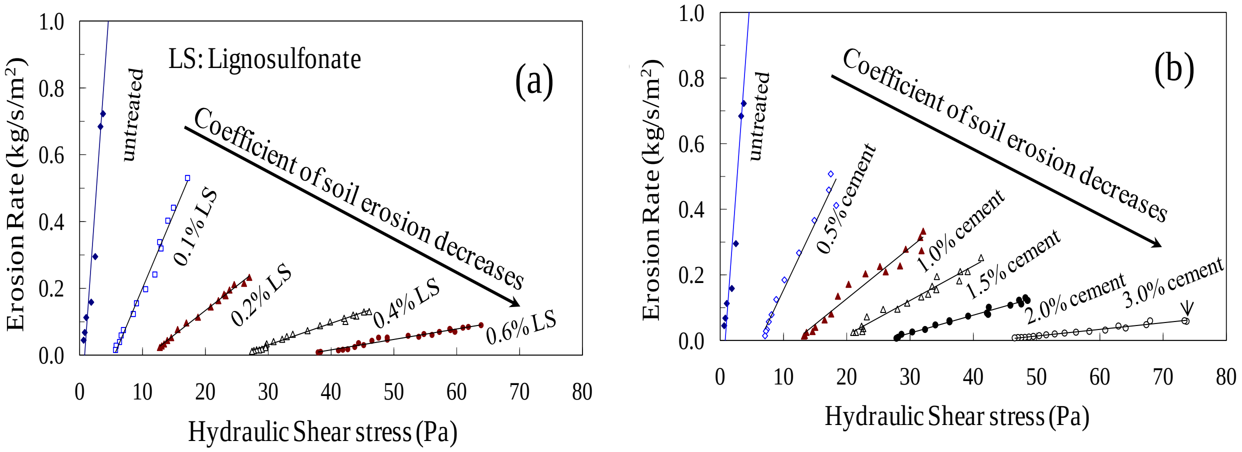


Figure 2: Erosion rate against hydraulic shear stress for (a) lignosulfonate and (b) cement treated soil (Indraratna et al., 2009)

4 EROSION MODEL FOR LIGNSOULFONATE TREATED SOIL

Indraratna et al. (2013) developed a rigorous theoretical erosion model to predict the erosion rate incorporating the shear strength characteristics of lignosulfonate treated soil. The energy required by the soil particles for mass erosion was formulated and equated to the energy dissipated by the fluid flow by adapting law of conservation of energy.

When the hydrodynamic forces placed on chemically bonded grains exceed a critical value, the inter-particle bonds are broken and the grains come into suspension. These suspended particles are transported by the eroding fluid, which completes the process of erosion. Therefore, energy required by the particles to erode should be equal to the energy dissipated by the excess hydraulic shear stress during erosion. This energy required by the particles to erode is the sum of the energy required to break the interparticle bonds and the energy required to bring the particle into full suspension reaching flow velocity, assuming no re-settlement occurs once the particles are fully suspended. The erosion model (Eq. 1) is based on the energy conservation principle and the formulation of aforementioned components. The full derivation of the proposed model (Eq. 1) can be found in Indraratna et al (2013). The erosion rate, (ε̇)*, of lignosulfonate treated soil can now be given as follows:

$$(\dot{\varepsilon})^* = \left[\frac{G_s \rho_w \omega V_f}{\frac{6q}{\pi D} A^* \left(1 - \frac{2}{3} \sin \phi'^* \tan \beta\right) + \frac{G_s \rho_w}{12} {V_f}^2}\right] (\tau_a - \tau_c^*) \qquad (1)$$

where, Gs and ρ_d are the specific gravity and dry density of soil, respectively; ρ_w is the density of eroding fluid; ω efficiency factor. The τ_a applied hydraulic shear stress and τ_c is critical shear stress initiating erosion; q and β are constants that depend on the packing arrangement of particles, D is the mean particle diameter, $(\emptyset')^*$ internal friction angle of treated soil; A^* is determined from the strain energy density of soil up to failure using the stress-strain curves; m is the mass of a single soil grain; V_t linear velocity of the moving soil particle in tangential direction; V_f is the mean flow velocity.

4.1 SHEAR STRENGHT PARAMETERS

In order to establish the shear strength parameters of the proposed erosion model, a series of consolidated drained direct shear tests were carried out on lignosulfonate treated and untreated soil specimens. The shear tests were conducted for specimens having the same moisture content (OMC) and dry density (95% MDD) conditions used in the erosion tests to ensure reliability between the shear and erosion tests. Three lignosulfonate dosages (0.2%, 0.6% and 1.2% by dry soil weight) and five effective normal stresses (5kPa, 10kPa, 15kPa, 22kPa and 42kPa) were selected to study the shear behaviour of the stabilized soil. The required amount of LS was mixed with water before adding into soil. The samples were statically compacted, cured for seven days and then saturated for 24 hours in the sample container prior to shearing using a displacement rate of 0.05mm/min. A typical test result for different LS dosages and vertical stress of 10 and 22kPa is shown in Fig 3. It can be observed that with the LS treatment, the shear strength of silty sand increased at a given effective normal stress. However, the stiffness only increased slightly due to treatment, and the change in ductility was negligible. Also, the volume changes showed more dilative responses after treatment.

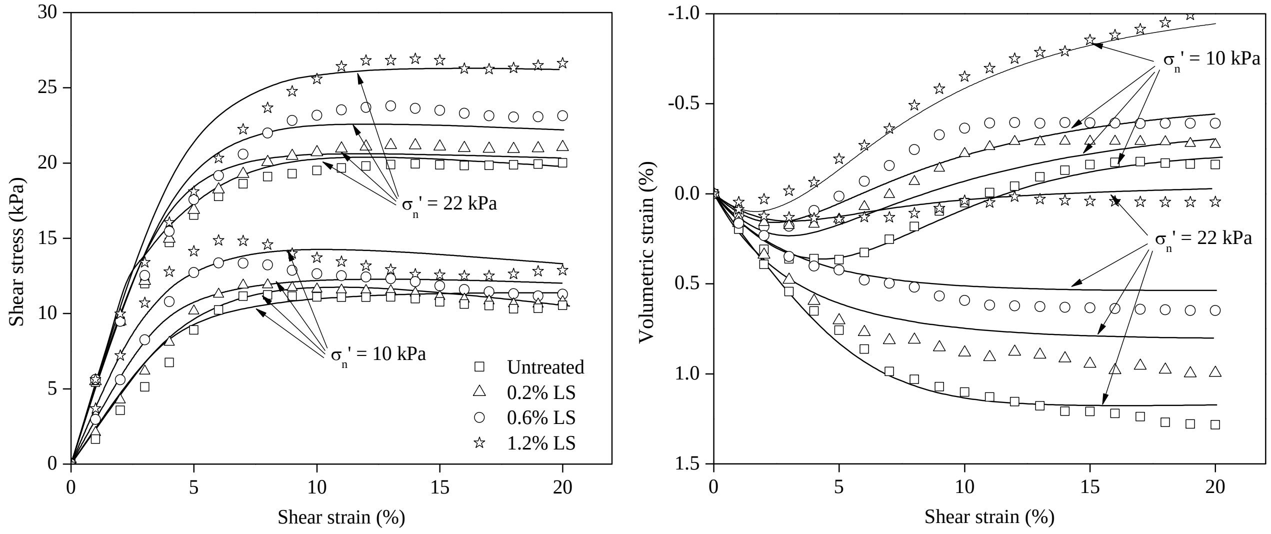


Figure 3: Stress-strain and volume change behaviour of lignosulfonate treated soil (after Athukorala et al 2015)

4.1 EROSION PARAMETERS AND MODEL VALIDATION

The proposed equation is validated with the laboratory experiments carried out using PSAICE for LS treated and untreated soils. A range of mean flow velocities were used to generate different hydraulic shear stresses for a given level of lignosulfonate treatment. Fig. 4 clearly shows the capability of Eq. (1) in predicting the erosion rate of soil for different amounts of lignosulfonate treatment over a wider range of hydraulic shear stresses. It should be noted that, the 0.1%, and 0.4% LS treated predictions are completely independent set of data as these experimental results were not used to determine the model parameters. The proposed erosion model can accurately capture the erosion of untreated soil and soil stabilised by lignosulfonate over a wide range of hydraulic shear stresses.

5 DSC MODEL FOR LIGNOSULFONATE TREATED ERODILBLE SOIL

The Disturbed State Concept (DSC) can be used to characterise the shear strength behaviour of the bonded materials such as cemented soils, as well as the untreated soil. However, there is no DSC model for bonded soils reported in literature. In this model, the relative intact (RI) responses of both treated and untreated soils were modelled incorporating the δ_0 version of the Hierarchical Single Surface (HiSS) plasticity models, whereas the non-associated yielding was considered through the disturbance function. The response of LS bonds was modelled using DSC considering a linear elastic RI response and zero strength condition as the relative states.

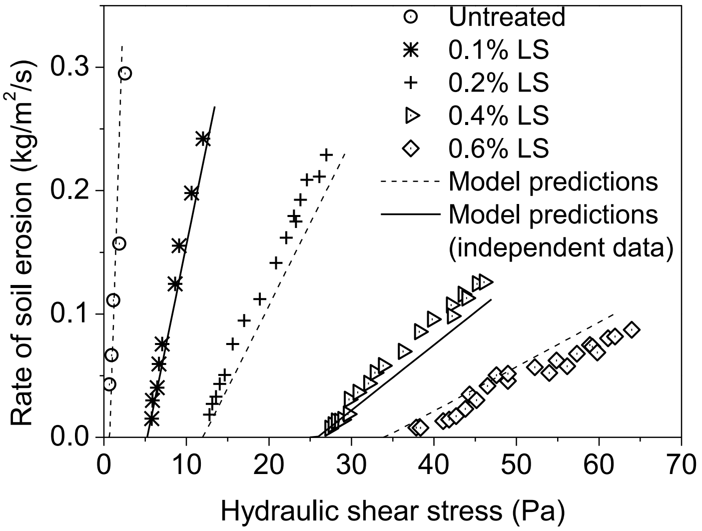


Figure. 4: Comparison of experimental and model predicted erosion rates (Indraratna et al 2013)

The reference states considered for this DSC model and the disturbance for lignosulfonate treated soils are illustrated in Fig. 5.

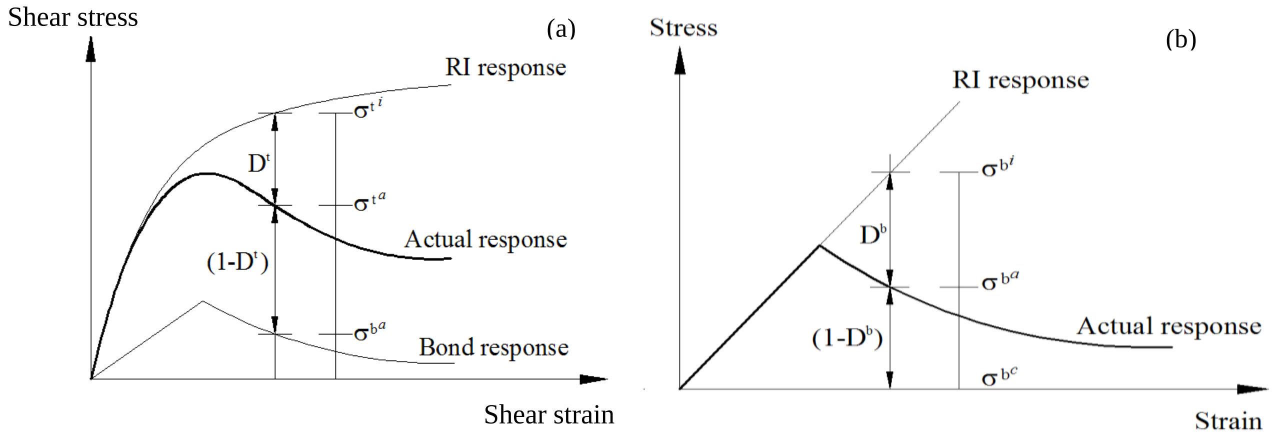


Figure 5: (a) Disturbance behaviour for lignosulfonate treated soil and (b) DSC for the lignosulfonate bonds (Athukorala et al 2015)

The incremental stress-strain relationship for the actual response of lignosulfonate treated soil can be evaluated as:

$$d\underset{\sim}{\sigma}^{t^a} = \underset{\sim}{C}^{t^{DSC}} d\underset{\sim}{\varepsilon}^{t^i} \qquad (2)$$

where $d\underset{\sim}{\varepsilon}^{t^i}$ is the vector of incremental strains of lignosulfonate treated soil in RI state and $\underset{\sim}{C}^{t^{DSC}}$ is the disturbed state concept constitutive matrix for lignosulfonate treated soil given by:

$$\underset{\sim}{C}^{t^{DSC}} = (1 - D^t)\underset{\sim}{C}^{t^{(ep)i}} + D^t \underset{\sim}{C}^{b^{DSC}} + \underset{\sim}{\sigma}_R^t \underset{\sim}{C}^{t^R} \qquad (3)$$

where, the disturbance function D^t, can be expressed in terms of the plastic strain trajectory ξ, as:

$$D^t = D_u^t \left[1 - e^{-A^t \left(\xi_D^t - \xi_D^{t*}\right)^{Z^t}} \right] \tag{4}$$

where D_u^t is the ultimate value of D^t at the residual, ξ_D^{t*} is the deviatoric plastic strain trajectory below which the disturbance is zero, and A^t and Z^t are the model parameters for lignosulfonate treated soil. In Eq. (3), $\underset{\sim}{\sigma}_R^t = \left(\underset{\sim}{\sigma}^{b^a} - \underset{\sim}{\sigma}^{t^i}\right)$, $\underset{\sim}{C}^{t(ep)i}$ is the elasto-plastic constitutive matrix for the RI behaviour of lignosulfonate treated soil, and $\underset{\sim}{C}^{t^R}$ is given by:

$$\underset{\sim}{C}^{t^R} = \frac{\left[D_u^t A^t Z^t \left(\xi_D^t - \xi_D^{t*}\right)^{(Z^t - 1)} e^{-A^t \left(\xi_D^t - \xi_D^{t*}\right)^{Z^t}} \right] \left[\left(\frac{\partial F^t}{\partial \underset{\sim}{\sigma}}\right)^T \cdot \frac{\partial F^t}{\partial \underset{\sim}{\sigma}} \right]^{1/2}}{\left(\frac{\partial F^t}{\partial \underset{\sim}{\sigma}}\right)^T \cdot \underset{\sim}{C}^{t(e)i} \cdot \frac{\partial F^t}{\partial \underset{\sim}{\sigma}} - \frac{\partial F^t}{\partial \xi_D^t} \cdot \left[\left(\frac{\partial F^t}{\partial \underset{\sim}{\sigma}}\right)^T \cdot \frac{\partial F^t}{\partial \underset{\sim}{\sigma}} \right]^{1/2}} \left(\frac{\partial F^t}{\partial \underset{\sim}{\sigma}}\right)^T \cdot \underset{\sim}{C}^{t(e)i} \tag{5}$$

The fully adjusted (FA) parameters for lignosulfonate treated soil, the experimental bond strengths were calculated from the laboratory shear test results as the difference between the shear stresses of lignosulfonate treated and untreated soil. More details on the model development can be found elsewhere (e.g. Athukorala et al. 2015)

The comparisons of the predicted stress-strain curves (solid lines) with the experimental data (bullet points) are also shown in Fig. 3. It can be observed that the proposed DSC model can predict with reasonable accuracy the experimentally trends obtained for the LS treated soil stress-strain behaviour. In addition, Fig. 3 also confirms that the predicted volume changes are in good agreement with the experimental observations.

6 EXPANSIVE SOIL STABILISATION USING LIGNOSULFONATE

The potential of use of Lignosulfonate (LS) in controlling the swelling of expansive soils is shown in Fig. 6. One dimensional swell tests were performed on untreated and LS treated remoulded samples on a typical Australian expansive soil collected in Queensland. The test results indicated that LS has significant role in controlling the swelling behaviour of this expansive soil (Fig. 6). For instance, as the LS content increases the long term vertical expansion is minimised (Fig 6a), however larger reduction is observed for small LS dosages, i.e. LS=2%. Furthermore, it seems as the effectiveness of LS in controlling swelling is more effective for low applied vertical pressures, i.e. <80kPa; after this value is exceeded the applied pressure governs the volumetric behaviour to a greater extent (Fig 6b). This vertical pressure range is consistent with that of typical transport infrastructure which indicates that LS is a viable stabilization alternative. Moreover, the magnitude of the swelling pressure of the soil was measured to be 105kPa but decreased to 84kPa upon 2% LS addition as shown in Fig. 6b. The swelling behaviour of identical specimens treated with a traditional admixture (cement) was also studied. For cement-treated specimens, the swelling pressure is similar to that of 2% LS (82kPa). This suggests that LS could be a resourceful alternative, considering its low cost and lesser environmental footprint comparatively to traditional alkaline additives for treatment of expansive soil in view of sustainable use of waste by-products and green construction.

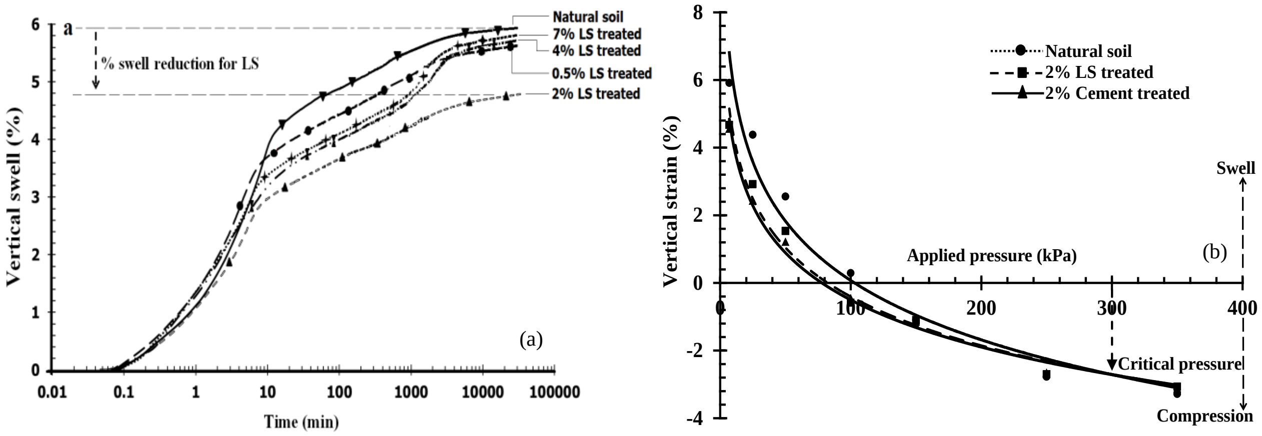


Figure 6: Swelling behaviour of an expansive soil treated with LS and cement for (a) a 7kPa seating vertical pressure and (b) variable applied vertical pressure (Alazigha et al., 2016).

6.1 MICROSTRUCTURAL ANALYSIS

XRD was employed in this study to assess the changes in clay mineralogy due to LS addition. The XRD diffractograms of the untreated and 2% LS treated soil are presented in Fig 7. The diffractograms were compared for the existence of new peaks (new minerals) in treated samples. The distance of the separation of atomic planes (d-spacing) was also investigated to understand the mineralogical alterations in treated specimen. A comparison of untreated and 2% LS treated diffractograms indicates a shift at the 001 montmorillonite peak to the left (an increase in d-spacing) for treated sample. This result suggests that LS intercalated the inner layer of the montmorillonite lattice and forced it to initially expand. In addition, the montmorillonite patterns either broadened or completely absent in the treated sample. The changes in the original montmorillonite lattice suggests that LS altered or destroyed the montmorillonite lattices. The peak intensities of other minerals such as quartz, kaolinite and illite also decreased. However, XRD patterns did not show intercalation of LS into the interlayers of these minerals. This suggests that only peripheral adsorption of LS by these minerals took place. This could be a result of the large molecular weight of LS polymer as compared to the interlayer spaces in the non-expansive minerals. Thus, the amorphous LS stabilizer underwent basal and peripheral adsorption on montmorillonite lattices and only peripheral adsorption on other soil minerals. The adsorption process subsequently coated the clay mineral lattices to form flocs and restricted the characteristic diffraction of the atomic planes especially in montmorillonite and also limiting the movement of water molecules into the entire clay matrix which in turn reduced swelling.

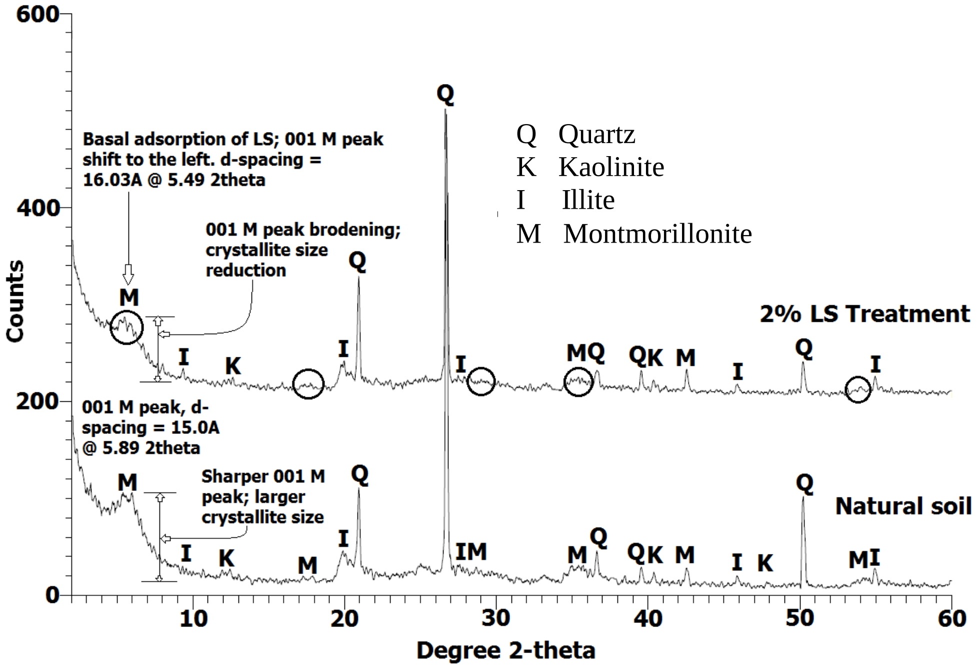


Figure 7: XRD diffractograms of untreated and 2% LS treated expansive soil (Alazigha et al., 2016**)**

The microstructure of the stabilized soil specimens was studied using scanning electron microscope (SEM) analytical technique. Fig 8 shows the micrographs of the untreated and the treated (2% LS) specimens at a magnification of 3000x. It can be observed marked changes in morphology, the untreated soil structure has transformed from a particle based form to a more integrated composite with larger but fewer inter-aggregates dominate pore spaces. Monroy et al. (2005) reported that the formation of larger inter-aggregate pore spaces causes reduction in the number and volume of intra-aggregate pore spaces which restrict the movement of water molecules into the entire clay matrix.

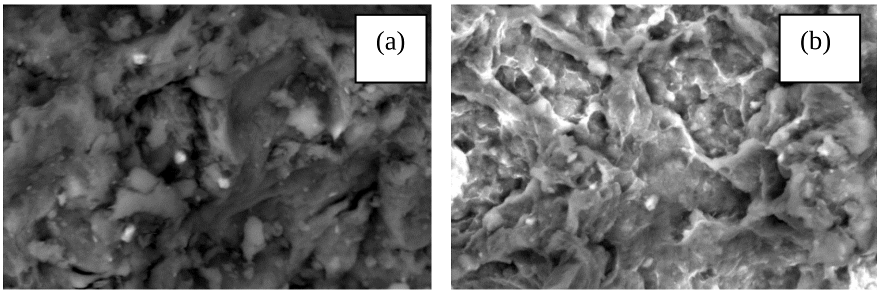


Figure 8: SEM micrographs (a) remoulded untreated (b) 2% LS treated expansive soil (Alazigha et al., 2016)

7 STABILIZATION MECHANISM

The stabilization process by lignosulfonate can be described by the schematic diagram as shown in Fig. 9. Fig. 9a shows a typical soil mineralogical structure, where clay mineral layers with excess negative charges is bonded by interlayer bonding materials (circle). Once the soil mass is treated with lignosulfonate, adsorption occurs on the clay mineral surface by electrostatic attraction (Fig. 9b). Then with time, lignosulfonate will neutralize the excess negative charges of the clay mineral surface resulting in the formation of bonding with clay particles (Fig. 9c). Finally, the lignosulfonate polymer chain draws the clay particles together to form an aggregate or grain cluster (Fig. 9d).

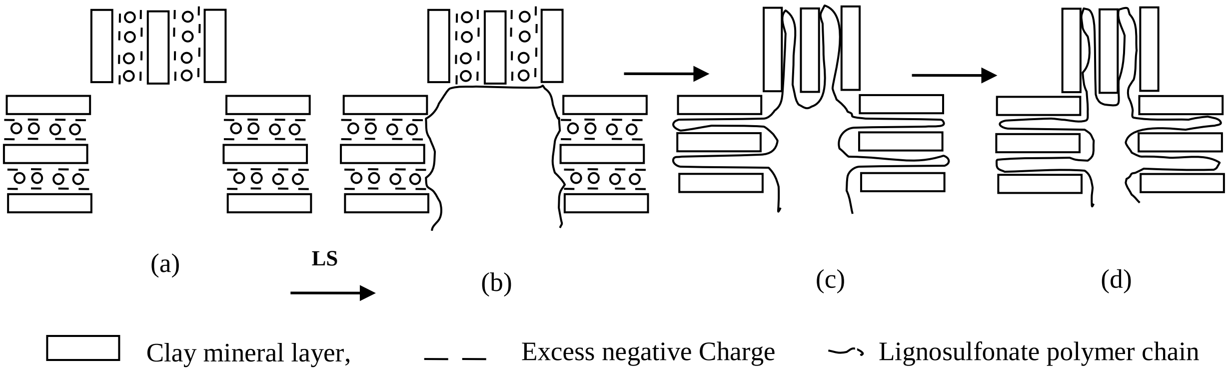


Figure.9: Schematic diagram for stabilization of soil by LS (a) typical mineralogical structure of clay (b) adsorption of LS on clay surface; (c) neutralisation of excess negative charges (d) LS polymer chain forming soil aggregate (Vinod et al., 2010)

8 CONCLUSIONS AND RECOMMENDATIONS

This paper presents the results of a study on the internal erosion behaviour of LS treated soil. Laboratory experiments carried out using PSAICE highlight that the lignosulfonate treatment reduces the coefficient of soil erosion significantly while increasing the critical shear stress required to initiate erosion.

The proposed erosion model can estimate the erosion rate over a wide range of hydraulic shear stress for different amount of LS similar to the laboratory experiments. This model can be adopted in practice to evaluate internal erosion in assessing the safety of earth structures such as rail/road embankments and dam cores.

A constitutive model for lignosulfonate treated silty sand was proposed, incorporating the original Disturbed State Concept. The LS bond strength was modelled separately using a linear elastic RI response and zero strength state as the reference states. A series of drained direct shear tests were carried out for a lignosulfonate treated silty sand and the test results were used to calculate the relevant model parameters. The model predicted stress-strain and volume change behaviour was compared with an independent set of experimental test results. The comparisons verified that the proposed DSC model can accurately capture the changes in shear strength, stiffness and the volume changes due to lignosulfonate treatment.

The amount of LS required to effectively control the swelling behaviour of the expansive soil is about 2% dry weight of soil and this value seems to be related to the stabilization mechanisms of LS. It was also observed that the soil pH does not change upon LS addition which is in direct contrast with cement admixture which considerably increased soil pH from 7.43 to 9.19 at an application rate of 2% by weight, and this may cause corrosion problems on buried steel infrastructure.

The values of free swell, swelling pressure were improved with the addition of LS admixture. The percent swell of 2% LS treated soil reduced about 2% after 7 days of curing and swelling pressure decreased from 105kPa to 84kPa. This implies that in the medium to long term, 2% LS treated expansive soils could be beneficial in improving the stability of treated foundations.

Micro-chemical analysis reveals that the improvement of performance exhibited by the lignosulfonate treated soil is attributed to the reduction of the double layer thickness by the neutralization of surface charges of the clay particles and the subsequent formation of a stable particle cluster or aggregate that will retain the ductile behaviour of the treated soil that is beneficial for sustainable infrastructure development compared to the enhanced brittleness often introduced by traditional chemical treatments.

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