Stabilisation of an erodible soil using a chemical admixture

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Abstract
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I. INTRODUCTION

Highly erodible and dispersive soils are common in many parts of Australia. These soils cover all states in Australia as top soils that have been affected by harsh climate change effects, leading to significant soil loss from large terrains, embankments, mass movement down slopes, surface and internal washout from earth dams and rapid erosion of canal banks. The change in the global climate has also resulted in the greater fluctuations of the temperature and lowering of the groundwater table causing considerable loss of interstitial moisture of surface soil particles, thereby losing inter-particle cohesion. Attributed to the lack of sufficient cohesion in the soil matrix, it becomes unstable causing rapid erosion when subjected to all forms of natural destabilising events (e.g. sudden heavy precipitation, high winds, flooding of low-lying areas). In Australia, soil loss by erosion exceeds several billion tonnes every year (Australian Agriculture Assessment, 2001). Rapid erosion of surface soil, internal washout and piping in embankments and dams containing dispersive and collapsible soils pose significant construction, stability and maintenance problems. Indeed, the prevention of soil loss and associated damage is currently a key element in the national priority goals of 'Environmentally Sustainable Australia' (ARC, 2008). Moreover, given the ever-increasing demand for infrastructure development that will encompass a greater extent of problematic soils, it is strategically important to improve the erosion and dispersive resistance of surface and shallow soil deposits in an appropriate, cost-effective and environmentally sustainable manner.

Chemical stabilisation is an effective ground improvement technique for controlling erosion. The traditional chemical stabilisers (e.g. lime, cement, gypsum and fly ash) are not readily acceptable, however, due to stringent occupational health and safety issues. Furthermore, they pose various threats to the environment by changing the soil and groundwater pH, which often affects the vegetation and subsurface native fauna. A large number of Australian species and grasses are threatened by pH values greater than 8.5. Moreover, the conventional cemented soils (often alkaline due to cement, lime and gypsum treatment) have shown brittle behaviour (e.g. Kamruzzaman et al., 2009; Sariosseiri and Muhunthan, 2009) especially under cyclic loads, affecting the stability of high-speed rail embankments and aircraft runways. Moreover, a decrease in the porosity and permeability can also occur due to conventional chemical treatment that contributes to reduced drainage (e.g. de Brito Galva et al., 2004; Desmet et al., 1985; Rajasekaran and Narasimha Rao, 1997). To overcome these difficulties, researchers are now looking for alternative soil stabilisers that will maintain sufficiently ductile soil properties, thereby preventing brittle (tensile) cracking. In the above context, lignosulfonate has shown a promising prospect as a stabilising agent especially for numerous erodible and dispersive soils in Australia (Indraratna et al., 2008). It belongs to a family of lignin-based organic polymers derived as a waste by-product from the wood and paper-processing industry. In comparison with highly alkaline and sometimes corrosive chemical admixtures, lignosulfonate is an environmentally friendly, non-corrosive and non-toxic chemical that does not alter the soil pH upon treatment. Due to the very small amount needed for effective soil treatment, there is also no leaching to affect the groundwater chemistry (e.g. Desmet et al., 1985).
In the recent past, investigations have been carried out on cohesive soils with lignosulfonate as a strength-improving stabiliser (Puppala and Hanchanloet, 1999; Tingle and Santori, 2003). It is reported that lignosulfonate with a very small amount of sulfuric acid as a catalytic additive showed a profound improvement in shear strength and resilient modulus of soil. Tingle and Santori (2003) investigated the effect of lignosulfonate on different clayey soils and they found that some types of lignosulfonates were able to significantly improve the strength of clayey soils of low plasticity. In addition, a number of researchers have performed experiments to investigate whether such lignin-based products if used in low volumes in road construction would improve the strength of the sub-grade and control surface dust emission (e.g. Chemstab, 2003; Tingle and Santori, 2003). Recently, Indraratna et al. (2008) conducted research on internal erosion behaviour of lignosulfonate-treated dispersive soils using a process simulation apparatus for internal crack erosion (PSAICE) designed and built at the University of Wollongong (UoW). They demonstrated that lignosulfonate treatment improved the erosion resistance of the treated soil similar to traditional admixtures such as lime and cement. However, the actual stabilisation mechanism of lignosulfonate-treated soil still remains questionable as they are not the typical cementitious admixtures that are commonly used in practice. The present study explains the probable mechanisms of stabilisation of lignosulfonate-treated soil based on micro-chemical analysis.

2. PROPERTIES OF LIGNOSULFONATE AS AN ENVIRONMENTALLY SUSTAINABLE ADMIXTURE

The main advantages of lignosulfonate over traditional stabilisers are non-toxicity, non-corrosiveness and environmental sustainability in promoting surface vegetation and natural subsurface fauna, which helps in retaining the soil carbon sequestration potential. The stabilisation mechanisms of traditional stabilisers mainly consist of cation exchange, pozzolanic reactivity and associated cementation by crystal growth in soil pores (gelation), and flocculation resulting in the formation of calcium silicate hydrates (C–S–H) and calcium aluminium silicate hydrates (C–A–S–H) cementitious compounds (e.g. Bell, 1996; Chew et al., 2004). Soil treated with traditional stabilisers such as lime, gypsum and cements also increased the pH of pore water to a value over 9 (e.g. Rollings et al., 1999). Field reconnaissance implied that the increase in the pH of groundwater to levels exceeding 9, for instance, due to excessive lime treatment may cause damage to certain species of native vegetation and to the aquatic habitats of nearby waterways. The increase in the soil pH can further influence the longevity of construction materials in the ground substructures (e.g. Biggs and Mahony, 2004; Perry, 1977) while presenting a threat to groundwater pollution in the land areas where groundwater is pumped out for drinking purposes. In the field experience of the authors, unlike the case of cemented soils upon conventional chemical treatment, the addition of lignosulfonate has been proved to maintain sufficient soil permeability (e.g. Desmet et al., 1985) and a water-retention capacity to supply the necessary nutrition for bio-engineering. The promotion of bio-engineering (e.g. native vegetation or green corridors) has a large effect on building up of the soil carbon level, which has a direct impact on the global climatic change. Studies have highlighted that soil contains approximately twice the amount of carbon in the atmosphere (e.g. Chan, 2008). Moreover, Shulga et al. (2008) observed that lignosulfonate develops a soil macrostructure that provides an excellent reinforcing effect to the soil matrix for withstand wind and water erosion of desiccated sandy soils, thereby ensuring relatively long-term stability. Lignosulfonate is a lignin-based polymer compound documented by researchers (e.g. Lemes et al., 2005; Mollah et al., 1995; Moustafa et al., 1981). It consists of both hydrophilic groups including sulfate, phenylic hydroxyl, alcoholic hydroxyl and hydrophobic groups including the carbon chain (e.g. Chen, 2004). According to Fredheim and Christensen (2003) the molecular weight of lignosulfonate varies from 4600 to 398 000 g/mol and these are soluble in water over the entire range of pH. As lignosulfonate is a non-toxic, non-corrosive chemical substance and can be used for the stabilisation of soil without producing any harmful chemical compounds, it can be considered as an environmentally friendly chemical stabiliser (Indraratna et al., 2008).

3. THEORETICAL BACKGROUND

The PSAICE was used to study the internal erosion behaviour of lignosulfonate-treated and untreated soils. The results of the experimental study are presented in the form of erosion rate with hydraulic shear stress. The erosion rate and hydraulic shear stress determined were then used to calculate the erosion parameters, namely the critical shear stress and the coefficient of soil erosion. The coefficient of soil erosion is the slope of erosion rate against hydraulic shear stress line and the critical shear stress, \( \tau_c \), is defined as the minimum hydraulic shear stress necessary to initiate erosion (Figure 1). The critical shear stress is estimated by extrapolating the straight line to the zero erosion rate and the slope of the linear line was presumed to be the coefficient of soil erosion (Indraratna et al., 2008). It was found that the turbidity increased initially, and then decreased as erosion progressed (Indraratna et al., 2008). However, the flow rate was observed to increase with time. The erosion rate, \( e \) (kg/s per m\(^2\)), can then be calculated using Equation 1 as described by Indraratna et al. (2008)

![Figure 1. Typical plot of erosion rate plotted against hydraulic shear stress](image-url)
where \( k \) (\( \text{kg/m}^3 \text{ per NTU (nephelometric turbidity unit)} \)) is the empirical factor relating turbidity to the soil solids concentrated in the flow, \( Q \) (\( \text{m}^3/\text{s} \)) is the average flow rate through the soil crack, \( T \) (NTU) is the average turbidity of the effluent, \( \phi_i \) (m) is the diameter of the soil crack at time \( t \), and \( l \) (m) is the length of the soil crack.

The hydraulic shear stress is determined using the friction factor method (Indraratna et al., 2008) as follows:

\[
\tau = \frac{f \rho w v^2}{8}
\]

where \( f \) is the friction factor; \( \rho_w \) (\( \text{kg/m}^3 \)) is the density of the eroding fluid; and \( v \) (m/s) is the mean velocity of the flow through the crack at time \( t \), which can be calculated using the flow rate and diameter of the crack. The friction factor was calculated from the Moody diagram (Abulnaga, 2002) based on the relative roughness and the Reynolds number. The relative roughness is calculated from Equation 3 as reported by Indraratna et al. (2008)

\[
\varepsilon = \frac{D}{2\phi_i}
\]

where \( D \) (m) is the mean particle diameter. The height of the roughness element was taken as the radius of the mean particle. The mean particle diameter was estimated from the particle size distribution of the eroded particles obtained using the Malvern particle size analyser. For dispersive clay, the mean particle diameter was observed to be 19, 22 and 25 \( \mu \)m for 0-2, 0-4 and 0-6% lignosulfonate treatment, and also 18, 28 and 32 \( \mu \)m for 0-2, 0-4 and 0-6% cement treatment, respectively.

The Reynolds number can be calculated using Equation 4

\[
R_e = \frac{\rho w v \phi_i}{\mu}
\]

where \( \mu \) (\( \text{kg/m per s} \)) is the dynamic viscosity of the eroding fluid.

### 4. LABORATORY TESTING

#### 4.1. Internal crack erosion tests

A series of internal crack erosion tests were conducted on dispersive clayey soil collected from Wakool, New South Wales (NSW), Australia. From the standard pinhole test (ASTM D4647) the dispersive clay is classified as D2, which represents highly dispersive clay that fails rapidly under 50 mm head (ASTM, 2004). The maximum dry density and optimum moisture content of the clay soil were found to be 15 kN/m\(^3\) and 22%, respectively. Furthermore, the liquid limit and plastic limit of the dispersive clay were found to be 47-6 and 29-4%, respectively. Various amounts of additive including lignosulfonate and cement ((0-2–0.6%), by dry weight of soil) were selected to stabilise the dispersive clay. General-purpose Portland cement manufactured in Australia and lignosulfonate were selected for this study. The lignosulfonate is completely soluble in water, a dark brown liquid having a pH value of approximately 4. This stabiliser is inflammable, it does not corrode metals, and it is classified as non-hazardous according to the National Occupational Health and Safety Commission (NOHSC) criteria (Chemstab, 2003). The selected soil was mixed with the selected amounts of lignosulfonate additive and statically compacted to 95% of the dry density inside a copper mould having dimensions of 72 mm diameter and 100 mm high. The prepared samples were wrapped in a moisture-proof bag and cured for 7 days. After curing, the samples were immersed in the eroding fluid (tap water) until saturation. Subsequently, internal crack erosion tests (PSAICE) were carried out by forcing the eroding fluid through a 10 mm soil crack (made by drilling with a guide block) at the centre of the samples.

The PSAICE equipment has an adjustable 25 litre tank applying the hydraulic gradients ranging from 0-5 to 7-0. The eroding fluid is stored in a 1000 litre tank and pumped into the moving constant head tank during testing. A photograph of the experimental set-up is shown in Figure 2. Two pressure transducers were connected to both ends of the sample to measure pressure difference across the crack. To measure the erosion rate continuously, an in-line process turbidity meter with an overall span of 0–3500 NTU is connected next to the downstream side of the soil sample to constantly monitor the effluent turbidity during the erosion test. The turbidity values were then used with the relationship developed between the concentration of solids (\( \text{kg/m}^3 \)) and turbidity (NTU) of the selected soil by Indraratna et al. (2008) to calculate the erosion rate. In order to continuously measure the flow rate, the effluent was weighed with an electronic balance. The pressure transducers, the turbidity meter, and the electronic balance are connected to a data acquisition system. More details on the test procedure can be found elsewhere (Indraratna et al., 2008).

### 5. RESULTS AND DISCUSSION

The variation of erosion rate with hydraulic shear stress for lignosulfonate-treated and untreated dispersive clay is presented in Figure 3. It is evident that the erosion rate and hydraulic shear stress follow a linear relationship and the slope represents the coefficient of soil erosion. As expected, critical...
shear stress increased and the coefficient of soil erosion decreased with the increase in the amount of lignosulfonate. As shown in Figure 3, the critical shear stress increased from 3.6 to 27 Pa with the addition of 0.6% lignosulfonate and the coefficient of soil erosion decreased from 0.019 to 0.0012 s/m.

The behaviour of lignosulfonate-treated dispersive clay has been compared with the cement-treated soils. General-purpose Portland cement was used for soil stabilisation and erosion tests were carried out on cement-treated soils very similar to lignosulfonate-stabilised soils. Figure 4 presents the variation of erosion rate plotted against hydraulic shear stress for cement-treated silty sand and dispersive clay. Critical shear stress increased and the coefficient of soil erosion decreased with the increase in the amount of cement (Figure 4). This behaviour was similar to that reported for lignosulfonate-treated soils. In addition, Indraratna et al. (2008) reported that the amount of lignosulfonate needed was much smaller in comparison with cement for similar gains in engineering behaviour for silty sand.

The performance improvement in terms of critical shear stress due to the addition of chemical additives (lignosulfonate and cement) can be represented as a non-dimensional critical shear stress ratio (CSSR), which is defined as the ratio of the critical shear stress of treated soil to the critical shear stress of untreated soil. The variation of critical shear stress ratio for lignosulfonate- and cement-treated dispersive clay is tabulated in Table 1. It is evident from Table 1 that for dispersive clay, the performance improvement in terms of CSSR was observed to be slightly better for cement-treated soil when compared with lignosulfonate.

### 6. MICRO-CHEMICAL ANALYSIS OF TREATED AND UNTREATED DISPERSIVE CLAY

Detailed knowledge about the clay particles and lignosulfonate interaction mechanism is very important for long-term environmental sustainability – a factor which is poorly understood at the micro-level. The micro-chemical analysis of treated and untreated dispersive clay was performed using X-ray diffraction (XRD) analysis, Fourier transform infrared resonance (FTIR) spectroscopy and scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS). The XRD analysis was performed to examine the formation of new compounds and crystalline size of clay minerals for lignosulfonate-treated and -untreated dispersive clay. For this purpose, XRD studies in the air-dried state were carried out for treated and untreated soil samples as described by Drits et al. (1997). The XRD result reveals that dispersive clay was composed of kaolinite, illite, montmorillonite and quartz (Figure 5). However, the XRD results did not exhibit any new peak due to amorphous or non-crystalline compounds (Figure 5). The crystalline sizes of the clay minerals were determined from the XRD data using Scherrer equation

$$ L = \frac{\lambda k}{\beta \cos \theta} $$

<table>
<thead>
<tr>
<th>Soil type</th>
<th>Amount of chemical: %</th>
<th>Critical shear stress ratio (CSSR)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Lignosulfonate</td>
</tr>
<tr>
<td>Dispersive clay</td>
<td>0.2</td>
<td>2.5</td>
</tr>
<tr>
<td></td>
<td>0.4</td>
<td>4.5</td>
</tr>
<tr>
<td></td>
<td>0.6</td>
<td>6.7</td>
</tr>
</tbody>
</table>

Table 1. Variation of critical shear stress ratio for different lignosulfonate- and cement-treated soils
where $L$ is the mean crystalline dimension in angstroms along a line normal to the reflecting plane, namely crystalline size; $k$ is a constant nearly unity; $\lambda$ is the wavelength of the X-radiation (1.54051 Å); and $\beta$ is the width of a peak at half height expressed in radians of $2\theta$.

The mean crystalline sizes of clay minerals measured from full width half maximum (FWHM) of XRD peaks are presented in Table 2. It can be seen from Table 2 that the percentage reduction of crystalline size varied with the type of clay minerals. The order of crystalline size reduction was found to be montmorillonite > illite > kaolinite. This emphasises that the reduction in clay mineral size depended on the clay structure and interstitial bonding of clay minerals. Moreover the constant peak position of treated and untreated clay minerals indicated that there was no change of crystalline orientation due to treatment by lignosulfonate. This demonstrates that crystalline size reduction occurred due to the reduction of surface negative charges, namely the decrease of surface negative charges of clay minerals by lignosulfonate.

Therefore, it appears that the electrostatic reaction process between clay minerals (negative charged) and positively charged lignosulfonate resulted in a reduction of the clay mineral surface charge and hence enhanced the bonding between clay minerals and lignosulfonate. However, to confirm the clay mineral–lignosulfonate bonding and clay mineral–lignosulfonate interaction, further chemical analysis is necessary and will be addressed in future.

FTIR analysis was performed to determine the functional groups of the lignosulfonate additive as well as for lignosulfonate-treated and untreated dispersive clay. The FTIR results are presented in the form of percentage transmittance plotted against the wave number. The percentage transmittance is determined by the ratio of the intensity of the transmitted beam ($I_t$) to that of the incident beam ($I_o$)

$$T = \frac{I_t}{I_o} \times 100\%$$

The FTIR results reveal that the lignosulfonate consisted of functional groups such as OH (3380 cm$^{-1}$), benzene ring (1650, 1509, 728 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 –OCH$_3$ group (1270 cm$^{-1}$) and S=O stretching sulfonate group (1184 cm$^{-1}$) as shown in Figure. 6. The chemical composition of lignosulfonate was determined by SEM–EDS analysis. The SEM–EDS results show that lignosulfonate was composed of carbon (C), oxygen (O), sulfur (S), and sodium (Na) (Figure 7). Based on this analysis the structure of the lignosulfonate was developed and it is presented in Figure 8.
The FTIR analysis was performed on treated and untreated dispersive clay soil to investigate their functional groups. Figure 9(a) shows that the dispersive clay consisted of functional groups such as Si–O, Al–O–H and OH. FTIR analysis was also conducted on lignosulfonate-treated dispersive clay (Figure 9). The FTIR results indicate that lignosulfonate-treated dispersive clay contained the functional groups of untreated dispersive clay plus the major functional groups of lignosulfonate (wave number ranging from 2000 to 1200 cm\(^{-1}\)). There is a clear difference in spectra observed in the FTIR results of treated and untreated clay (Figure 9). The additional spectra observed in the FTIR results of lignosulfonate-treated clay in wave numbers ranging from 2000 to 1200 cm\(^{-1}\) confirm the presence of functional groups such as benzene (1650, 1517 cm\(^{-1}\)) and the CH group (1460 cm\(^{-1}\)). The presence of the functional groups of lignosulfonate in lignosulfonate-treated clay confirms the formation of ionic bonding between the clay mineral lattices and the lignosulfonate functional groups and also the existence of lignosulfonate in the interlayer spacing of clay minerals.

![Figure 6. FTIR pattern of lignosulfonate](image1)

![Figure 7. (a) SEM image of lignosulfonate; (b) EDS result of lignosulfonate](image2)

![Figure 8. Structure of lignosulfonate](image3)
7. PROPOSED STABILISATION MECHANISM OF LIGNOSULFONATE-TREATED DISPERSIVE CLAY

The mechanism for lignosulfonate treatment on the stabilisation of the dispersive clay has been interpreted based on micro-chemical analysis. When the soil is treated with lignosulfonate and water, the lignosulfonate first undergoes hydrolysis, and then disintegrates into hydrogen ($H^+$) and hydroxyl ions ($OH^–$) as shown in Figure 10(a). Thus lignosulfonate is protonated by hydrogen ions (Figure 10(b)). At this stage, the oxygen atom of a secondary hydroxyl will be protonated because it contains two pairs of unshared electrons in the outermost energy layer. On the other hand, the oxygen atom also contains a methoxyl ($–OCH_3$) radical. However, this oxygen atom would not associate with hydrogen bonding, because the oxygen of this radical bears a partially negative charge. Finally, the protonated lignosulfonate forms a positive-charged compound after releasing water (Figure 10(c)). The newly formed positive-charged lignosulfonate will neutralise the negative charges of the clay mineral surface because of electrostatic attraction. This electrostatic reaction (charge neutralisation reaction process) leads to the reduction of double-layer thickness (i.e. crystalline dimension) and the formation of bonding with clay minerals. The clay mineral–lignosulfonate interactions were confirmed by FTIR and the reduction of clay mineral size can be confirmed by XRD analysis. Lignosulfonate has a longer polymer chain which draws the clay particles together and forms aggregates. Therefore, the lignosulfonate–clay mineral intercalated compounds exhibit higher erosion resistance when compared with untreated dispersive clay.

The stabilisation process by lignosulfonate is described by the schematic diagram shown in Figure 11. A typical soil mineralogical structure, in which clay mineral layers with excess negative charges (dash) were bonded by interlayer bonding materials (circle) are shown in Figure 11(a). Once the soil mass has been treated with lignosulfonate, adsorption occurs on the clay mineral surface by electrostatic attraction (Figure 11(b)) and over a period of time the lignosulfonate will neutralise the excess negative charges of the clay mineral surface resulting in the formation of bonding with clay particles (Figure 11(c)). Finally, the lignosulfonate polymer chain draws the clay particles together to form an aggregate or grain cluster (Figure 11(d)).

8. CONCLUSIONS

Lignosulfonate is an environmentally sustainable admixture which stabilises the soil effectively while avoiding adverse environmental consequences such as significant changes in soil pH, groundwater pollution by leaching, and associated impacts.
on native vegetation and aquaculture. The results of the present study show the potential of lignosulfonate for stabilising dispersive soils against internal crack erosion. It was found that lignosulfonate would reduce the coefficient of soil erosion and significantly increase the critical shear stress of both clayey and silty soils. The enhancement of critical shear stress was expressed in terms of the CSSR. It is found that the stabilisation of the dispersive clay was more effective with 0.6% cement than 0.6% of lignosulfonate, although the soil improvement with the latter was still significant in view of the enhanced stress–strain behaviour. Micro-chemical analysis reveals that the improvement of performance exhibited by the lignosulfonate-treated soil can be attributed to the reduction of the double layer thickness by the reduction of the surface charges of the clay particles and the subsequent formation of a stable particle cluster or aggregate. This will retain the ductile behaviour of the...
treated soil which will be beneficial for sustainable infrastructure development compared with the enhanced brittleness often introduced by traditional chemical treatments. However, this proposed mechanism will be further investigated with the support of additional chemical analysis in future.

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