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Study on pollution of soils due to disposal of pulp mill effluent

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Abstract
Rapid industrialization resulted in the generation and disposal of large volumes of solid, liquid and gaseous wastes into the environment. Geotechnical characterization of soils polluted by industrial wastes is an essential aspect of Environmental Geotechniques. The present paper investigates the effect of pulp mill waste on volume change behavior of Bentonite and Kaolinite.Electrolyte concentration of the effluent emerges as the single most important factor influencing the interaction. The considerable increase in the permeability of bentonite treated with the effluent indicates that the extent of pollution would increase to a wider area with time affecting the structures over such soil. The increase in permeability also results ultimately in pollution of ground water resources.The significant decrease in permeability of kaolinite treated with the pulp mill effluent indicates possible drainage problems and consequent pounding up of the effluent at the disposal site, increasing the waste land formation. The paper presents the results of this study.

Keywords: Paper mill effluent, bentonite, kaolinite, permeability, environmental geotechniques, pollution, water resources

INTRODUCTION

Foundations to various Civil Engineering structures are generally designed based on the bearing capacity and settlement characteristics of the soil at the site prior to construction. Modification of soil properties subsequent to construction due to industrial pollution may cause unanticipated settlements or heaving and possible strength loss, which are not accounted for in the design, resulting in structuraldistress and consequent failure. Consideration for Environmental Stability of the foundation soil has therefore become an additional responsibility of the geotechnical engineers. Environmental Geotechniques is realized to be more of a planning and decision tool to be used for forecasting geotechnical problems that may ultimately arise, rather than solving after the event has occurred (Sembenelli&Ueshita, 1981). Such a requirement calls for a fore-knowledge of the effect of industrial effluents on the geotechnical properties of soil.

Case histories of foundation failures, structural damage in light industrial buildings on soils contaminated by various chemical pollutants have been reported. This emphasizes the importance which needs to be attached to the consideration of the above problem. In conventional Soil Mechanics, it is assumed that soil parameters such as liquid limit, Plastic limit etc., are constant for a given soil. However these parameters, far from being constant, change rapidly as pore fluid concentration and type changes. The present paper investigates the effects of effluent from the pulp mill section of a Paper mill in India on the Volume change behavior of bentonite and kaolinite.

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REVIEW OF RELATED LITERATURE

While enormous work has been done in different parts of the globe on the transport mechanisms of contaminants through the saturated and partially saturated soils and also on the ground water contamination, very few references are available on the effect of contaminants on the geotechnical properties of the soil. Lukas et al (1972) reported settlement of structure due to the chemical attack on soil. The settlement was attributed to the reduction of strength properties of the contaminated soil. Sridharan et al. (1981) reported the heaving of soil under the floors, beams and upheaval of foundations due to acid contamination. Soil with high phosphate content under acidic environment became the sources of the contamination. Such heaves lead to the distress of structure. Joshi et al (1994) reported the formation of calcium hydrogen phosphate hydrate when soil dominated by calcite and dolomite came in contact with the phosphoric acid. The resultant increase in molecular volume cause subsoil heave and differential movement which resulted damage to structures seated on such soil. In addition, some of the studies carried out in this area area are Kumapley (1985), Krov (1989), Srivastava et al(1994), NarasingaRao et al (1995), Cyrus et al (2010),Talukdar&Saikia (2010)etc.

MATERIALS AND METHODS

Materials

Commercially available bentonite and kaolinite have been used in the study. These two clays have been chosen for study, as they represent the two extreme limits of physic-chemical interaction with pollutants.Table-1 presents the gradationof the soils used for the study. The effluent collected is the black liquor produced as a waste from the pulp mill of M/s Coastal Papers Ltd., Kadiyam, Andhra Pradesh, India, having an installed capacity of 12,000 tons /annum of paper and license capacity of 33,000 tons/ annum. The black liquor is not treated either for chemical recovery or for disposal but conveyed through pipes and disposed off in a 20 hectares of land which is about 5 km from the plant. The characteristics of black liquor are shown in Table-2.

Table 1. Gradation of Soils used

Soil fraction	% of soil fraction	
	Bentonite	kaolinite
Coarse sand	0	0
Fine sand	5	20
silt	10	35
clay	85	45

Table 2. Characteristics of Pulp mill Effluent (Black liquor)

S. No	Property	Value
1	pH	10.5
2	Flow	1000 m^3/day
3	Colour	Black
4	Total solids	15,000 ppm
5	Suspended solids	4,500 ppm
6	COD	45,000 ppm
7	BOD	30,000 ppm
8	Organic matter	70,000 ppm
9	Sulphates	35,000 ppm
10	Treatment	nil

Treatment of soil with the Industrial effluent

The effluent from the industry was collected from the outlet of the plant in two 50 liter containers and brought to the laboratory. Sample of the effluent was retained for chemical analysis. Bentonite and kaolinite, in powder form, were added to the effluent in separate containers, accompanied by thorough mixing. A soil: effluent ratio of 1:15 was used. The treatment of the soils with the effluent was continued for 15 days, during which the soils were continually agitated. At the end of treatment period, the excess effluent was discarded.

The soil slurry so obtained was directly used for compressibility and sediment volume in distilled water. Part of the soil slurry was air dried, pulverized and passed through 425 μ sieve and the effluent treated dry soil powders were used for determination of liquid limit, plastic limit, specific gravity and sediment volume in carbon tetrachloride and for chemical and mineralogical tests.

Exchangeable cation Analysis

The exchangeable cation analysis was carried out by displacing the exchangeable cations of the untreated and effluent treated bentonite and kaolinite soil specimens. The cation exchange capacity (CEC) expressed as mille equivalents (m eq) per 100g of dry soil was computed from the sum of the individual exchangeable cations present. Table-3 presents the physic-chemical properties of untreated and pulp mill effluent treated soils.

Table 3. Physico-chemical properties of soils

S. No.	Soil	pH	Exchangeable Cations (meq/100 g)				
			Ca^{+2}	Mg^{+2}	Na^{+}	K^{+}	CEC
1	Untreated bentonite	7.5	28.3	3.90	47.1	0.80	80.1
2	Pulp mill effluent treated bentonite	10.5	29.6	4.10	56.5	0.20	90.4
3	Untreated kaolinite	7.9	8.40	0.72	4.30	0.21	13.63
4	Pulp mill effluent treated kaolinite	10.5	7.90	0.64	13.4	0.20	22.14

Index Properties

The liquid limit of the soils was determined by using the cone penetration method as per SP: 36 (1987a). Particle size distribution in the untreated soils was obtained by sieving coupled with hydrometer analysis as per SP: 36(1987b).

Because of the possible changes in the soil properties on drying, the sediment volume of the effluent treated soils was determined using un-dried soil slurries. The soil slurry, at an approximate water content of about twice the liquid limit was directly poured into a 100ml glass measuring jar, which was then filled up to 90ml mark with distilled water. The soil suspension was then thoroughly mixed using a glass rod and then the jar was filled up to the 100ml mark. When soil reached equilibrium volume in the measuring jar, as indicated by the negligible changes in the readings in two successive days, the final reading of sediment volume was taken. The excess water was carefully removed using a wash bottle with suction, the soil was carefully collected into a container and its oven dried weight was determined using an electrical balance having a sensitivity of 0.0001 g. Sediment volume was determined using the relation

$$FSI = \frac{V_d}{W_d} cm^3 / g$$

Oedometer Tests

An important consideration in the present investigation is i) to ensure effective interaction of the soil with the effluent and ii) to eliminate the effects of drying on the volume change behavior of untreated and effluent treated soils. Hence the method of Mesri& Olsen (1971), slightly modified to minimize or eliminate sample disturbance was adopted for the present study.

The effluent treated soils in the form of slurries, at a moisture content of about twice the liquid limit,were poured directly from the effluent treatment containers into the consolidation cell. The cell is initially assembled with bottom porous disc, filter paper and the consolidation ring. The pore fluid, which is the effluent, was allowed to drain through the bottom porous disc under no load. When the outflow from the bottom of the consolidation cell ceased, the top porous disc with filter paper was placed in position. When the drainage of the pore fluid due to load from the top porous disc ceased, the pressure pad and the loading ram are placed over the top porous disc and the dial gauge is assembled in position. The initial dial gauge reading is noted. The seating load of 5 kPa is applied after equilibrium is reached in the dial gauge reading.

Influence of Pulp Mill Effluent on Volume Change Behavior of Bentonite

Liquid Limit

Table-4 presents the liquid limit of untreated and pulp mill effluent treated bentonite. Treatment of bentonite with the effluent reduces the liquid limit considerably from 375% to 129%. The liquid limit of bentonite is known to vary directly with the thickness of the diffuse double layer surrounding the clay particles in an aqueous environment, the change in double layer thickness being brought about due to modified chemical environment. Important parameters influencing the double layer thickness are the cation valence and size, pH, dielectric constant and electrolyte concentration of the pore fluid and anion adsorption.

Table 4. Index Properties of Soils

S. No.	Soil type	Liquid Limit (%)	Plastic Limit (%)	Plasticity index	Free Swell Index	Shrinkage limit (%)
1	Untreatedbentonite	375	42.5	332.5	21.0	5
2	Effluent treated bentonite	129	68.4	60.6	14.2	14
3	Untreated kaolinite	30	19.2	10.8	1.4	20
4	Effluent treated kaolinite	32.5	20.0	12.5	2.0	28

It is observed that the changes in the exchangeable cations due to effluent treatment are marginal except for an increase in the exchangeable sodium. The increase in the exchangeable sodium should contribute to an increase in the double layer thickness and hence should increase the liquid limit, which is not true in the present case.

Chemical analysis of the effluent indicated significant amount of sulphates present in the effluent. The sulphate adsorption increases the liquid limit significantly (Sridharan et al, 1990). In the present case, effluent treatment caused a decrease in the liquid limit of bentonite and hence anion adsorption does not appear to have influenced the bentonite-pulp mill effluent interaction. The pH of the effluent is 10.5, which causes increase of negative surface charge and increase in double layer thickness (Taylor, 1959), which should result in an increase in liquid limit. The liquid limit in the present case decreases due to effluent interaction in spite of an alkaline environment. This suggests that the pH of the effluent did not influence the interaction.

Effect of Electrolyte Concentration

The pulp mill effluent, being a strong chemical contaminant possesses higher electrolyte concentration, than pore water. The high electrolyte concentration of the effluent suppresses the diffuse double layer causing a decrease in the liquid limit. Presumably, the influence of cation valence, pH and sulphate absorption is masked by the high electrolyte concentration of the effluent. Earlier studies on a montmorillonite soil showed that at high electrolyte concentration, the role of valence of exchangeable ion is negligible and the behavior is essentially controlled by electrolyte concentration (Sivapullaiah et al, 1994).

It is pointed out earlier that the effluent treated soil samples have not been washed with distilled water to represent more closely the field condition. To verify the influence of electrolyte concentration, the effluent treated bentonite has been subjected to a single washing with distilled water, followed by slow sedimentation and removal of the supernatant liquid. The liquid limit of the effluent treated bentonite specimen has been determined after washing with distilled water. Washing of effluent treated bentonite increased its liquid limit from 129% to 163%. Washing of effluent treated bentonite decreases the electrolyte concentration of the pore fluid, thus increasing the liquid limit.

Effect of drying

Liquid limit is known to be significantly reduced on drying and the causes of reduction are noted to be the presence of halloysite mineral, hydrated sesquioxides, carbonates and organic matter (Sridharan et al, 1989). While the presence of the first three is remote in the present case, the high BOD of the pulp mill effluent points to the possibility of reduction in liquid limit on drying due to presence of organic matter.

To verify the effect of drying, the liquid limit of effluent treated bentonite is determined after air-drying and oven-drying the effluent treated clay specimens. It is observed that the liquid limit of oven-dried effluent treated bentonite(97%) is significantly less than that of air-dried specimen (129%), indicating significant influence of drying.

The influence of drying verified above underlines the need to avoid drying in soil-effluent interaction studies. The procedure adopted for consolidation tests in this study is in line with this consideration.

Plastic Limit

Treatment of bentonite with the effluent increases its plastic limit significantly from 42.5% to 68.4%. The plasticity index decreases considerably from 332.5 to 60.6.

The decrease in double layer thickness and the associated repulsive forces due to effluent treatment contributes to a net increase in shear strength at particle contact. The higher shear strength at particle contact presumably requires greater amount of water to remold the effluent treated bentonite spacemen into thread corresponding to plastic limit water content.

Free Swell Index

Treatment of bentonite with the effluent decreases the free swell index considerably from 21 to 14.2 cm^3/g. The reduction of free swell volume of effluent treated bentonite is an evidence for the decrease in the thickness of diffuse double layer surrounding the bentonite particles. The decrease in double layer thickness decreases the inter-particle repulsive forces resulting in closer spacing of particles and lower free swell volume.

Compressibility Characteristics: Void ratio-Pressure relations

Fig.1 presents the void ratio- pressure relation for untreated and pulp mill effluent treated bentonite. The pulp mill effluent treated bentonite shows appreciably lower equilibrium void ratio at any pressure than the untreated bentonite. The e-log p curves of the untreated and effluent treated bentonite converge at a pressure of about 400kPa. It is well established (Mesri and Olsen, 1971) that the equilibrium void ratio mobilized by expanding lattice clays at a given pressure increment is a direct function of the physic-chemical repulsive forces arising from interaction of adjacent diffuse double layers. Treatment of bentonite clay specimen with the pulp mill effluent causes a reduction in the diffuse double layer thickness and the associated repulsive forces, resulting in a lower equilibrium void ratio at any pressure increment.

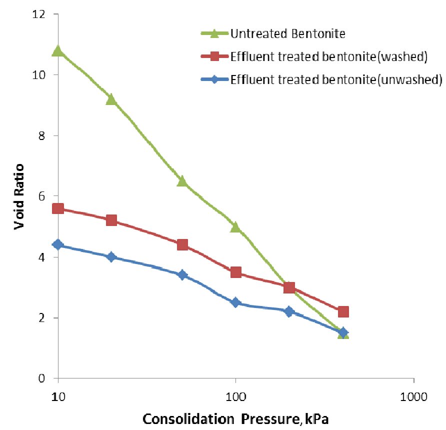

Fig 1. Effect of Effluent and its Electrolyte concentration on Pressure-Void Ratio relation for bentonite

Permeability- Pressure Relation

Fig 2 shows the permeability of untreated and pulp mill effluent treated bentonite as a function of applied pressure. It was observed that the permeability of contaminated soils can be better represented as a function of applied pressure than as void ratio (NarasingaRao et al, 2011). The effluent treated bentonite shows consistently higher permeability than the untreated bentonite at all consolidation pressures. The permeability increases by about 400 to 650% at a pressure of 10kPato 50 kPa and about 200 to 300% at a pressure of 100 to 400kPa due to effluent treatment. It has been brought out that the thickness of diffuse double layer reduces due to treatment with the pulp mill effluent resulting in increasing the effective void space available for fluid flow, which increases in permeability.

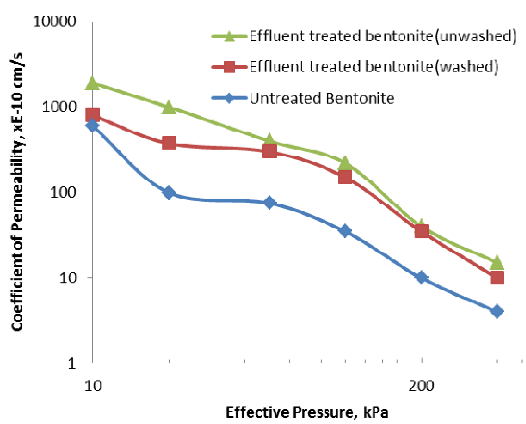

Fig 2. Effect of Pulp mill effluent treatment of permeability of bentonite

It may be noted that the reduction in the double layer thickness and the associated inter-particle repulsive forces results in a decrease in the inter-particle spacing and reduced permeability. Presumably, the increase in the effective void space due to reduction in the double layer thickness offsets the decrease in void ratio due to the reduction in inter-particle spacing, leading to a net increase in permeability. Further, the decrease in double layer thickness and the associated repulsive forces causes a more random arrangement of particles that reduces the tortuosity of flow path (Sridharan et al, 1986), leading to increase in the permeability.

Evidence to the effect of electrolyte concentration on compressibility

Fig. 1 presents the pressure void ratio curves of untreated bentonite as well as pulp mill effluent treated bentonite with and without washing with the distilled water. The effluent treated benonite washed with distilled water exhibits higher equilibrium void ratio than the unwashed effluent treated bentonite at any pressure. Washing of effluent treated bentonite decreases the electrolyte concentration of its pore fluid, which increases the thickness of double layer and the associated repulsive forces resulting in higher equilibrium void ratio for the effluent treated bentonite washed with distilled water than its unwashed counterpart at any pressure.

This provides an indirect evidence for the effect of the electrolyte concentration on the pulp mill effluent-bentonite interaction. Treatment of bentonite with the effluent increases its electrolyte concentration which is partly responsible for the lower equilibrium void ratio of the effluent treated bentonite than that of untreated bentonite. Washing of effluent treated bentonite partly reverses this process and decreases the electrolyte concentration thus shifting the pressure-void ratio curve towards that of the untreated bentonite.

Influence of Pulp mill effluent on volume change behavior of Kaolinite

Liquid Limit & Shrinkage limit

The liquid limit and shrinkage limit of the untreated and pulp mill effluent treated kaolinite is shown in Table-4. Treatment with the effluent increases the liquid limit of kaolinite marginally from 30% to 32.5%. The shrinkage limit of kaolinite increases significantly from 20% to 28% on treatment with the effluent. Sridharan and Rao (1975) brought out the mechanisms controlling the liquid limit of kaolinite. It was pointed out that for fixed lattice clays like kaolinite, the effect of physic-chemical forces are less significant than that of the fabric in influencing the liquid limit. A change in dielectric constant brings about more significant changes in the shearing resistance at particle contacts than in the diffused double layer.

The effect of electrolyte concentration of the effluent is to cause an apparent decrease in the dielectric constant of the effluent (Hasted et al, 1948). Thus for kaolinite, the higher electrolyte concentration of the pulp mill effluent, presumably causes an edge to face particle flocculation due to decrease in apparent dielectric constant. Further, pulp mill effluent, being a chemical contaminant, possesses a lower dielectric constant than water. The reduction in dielectric constant, due to treatment with pulp mill effluent, increases the shearing resistance at particle contact increasing the liquid limit. The resulting edge-to-face particle association offers more resistance to shrinkage causing an increase in the shrinkage limit.

Plastic limit & Plasticity index

The plastic limit of kaolinite increases from 19.2% to 23.7% after treatment with effluent (Table-4). The plasticity index increases from 10.8 to 16.3. The increased shear strength, due to treatment with the pulp mill effluent, presumably, requires greater amount of water to remold the clay into soil thread corresponding to plastic state, leading to increase in the plastic limit.

Void Ratio-Pressure Relations

Fig.3presents the void ratio-pressure relation for the untreated and pulp mill effluent treated kaolinite. It is observed from Fig.3 that the effluent treated kaolinite exhibits consistently higher void ratio than that of the untreated kaolinite at any pressure. The enhancement of particle flocculation due to effluent treatment resists the external pressure at higher void ratio.

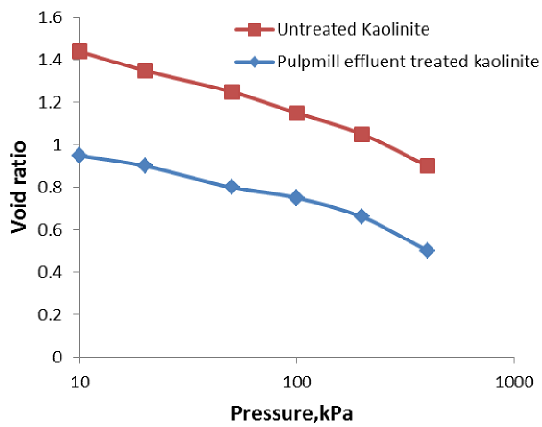

Fig 3. Effect of Pulpmill Effluent Compressibility of Kaolinite

Permeability–Relation pressure

Fig.4 presents the permeability versus consolidation pressure relation for natural and pulp mill effluent treated kaolinite. The permeability of effluent treated kaolinite decreases to of 14% of its untreated value at a pressure of 10kPa and to 50% at 400kPa. In spite of enhanced particle flocculation due to effluent treatment, as evident from the increased liquid limit, shrinkage limit and free swell index, the clogging of pores by the suspended solids present in the pulp mill effluent may be responsible for lower permeability of the effluent treated kaolinite than its untreated counterpart at any pressure.

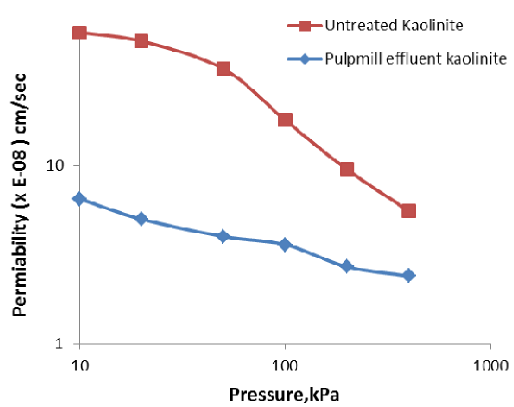

Fig 4. Effect of Pulpmill Effluent Permeability of Kaolinite

For bentonite, the effect of clogging of pores by dissolved solids has been presumably masked by the drastic reduction double layer thickness resulting in more effective void space available for fluid flow and hence higher permeability. It may be noted that the void ratio of bentonite is 4 times that of kaolinite at any pressure.

Consequences of Soil Industrial Effluent Interaction

1. Treatment of bentonite with pulp mill effluent resulted in a significant decrease in the liquid limit and equilibrium void ratio at any pressure increment. This indicates possible unexpected settlements to the existing structures over clayey soil deposits (containing montmorillonite clay mineral), which would cause distress to these structures.
2. The considerable increase in the permeability of bentonite treated with the effluent indicates that the extent of pollution would increase to a wider area with time affecting the structures over such soil. The increase in permeability also results ultimately in pollution of ground water resources.
3. The significant decrease in permeability of kaolinite treated with the pulp mill effluent indicates possible drainage problems and consequent pounding up of the effluent at the disposal site, increasing the waste land formation.

CONCLUSIONS

1. The treatment of bentonitewith pulp mill effluent decreases double layer thickness resulting in a decrease in liquid limit and free swell index and an increase in plastic limit and shrinkage limit of bentonite.
2. The effluent treated bentonite also showed lower equilibrium void ratio at any pressure, lesser compression, higher consolidation coefficient and higher permeability than its untreated counterparts.
3. For kaolinite,treatment with the pulp mill effluent caused a marginal increase in liquid limit and significant increase in free swell index and shrinkage limit.
4. The effluent treated kaolinite showed higher equilibrium void ratio at any pressure,but lower consolidation coefficient and permeability than its untreated counterparts.
5. Continual exposure of soils containing montmorilloniteand kaolinite minerals to the effluent pulp mill is likely to cause distress to existing or new structures over such soils, apart from endangering the environment.

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