Soils as compacted clay liners (CCL): raw material selection parameters

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Abstract: Ground and surface water are resources which should be protected from urban, agricultural and industrial pollution. Compacted Clay Liners (CCL) provide an alternative for landfill liners built in Brazil, because some regions have clayey sediments and clay stone. São Paulo state is undergoing important industrial development and 36 million people live there.

The rock formations of Paraná Basin constitute an important groundwater resource dominated by the Guaraní Aquifer outcrop in part of São Paulo State. The Paraná basin is composed in São Paulo of Devonian to Jurasic sedimentary rocks and extrusive Cretaceus rocks. These rocks are overlain by Cenozoic sediments which provide different raw materials for use as Compacted Clay Liners.

Different soils from the Corumbataí Formation were used in this study and were selected on the basis of their grain size distribution and clay mineralogy. The physical properties of the soils were determined using characterization tests. The clay minerals were determined by differential thermal analysis and X ray diffraction. Physico-chemical characterization was used to assess the competency of the material for use as liners. Batch equilibrium testing involving heavy metals was conducted to determine the adsorption isotherms.

The results of the physico-chemical analysis, mineralogical composition and adsorption isotherms indicate that materials from the Corumbataí Formations could be used as CCL.

Résumé: L'eaux, souterraines et superficielles, sont des ressources qui devraient être protégées de la pollution urbaine, agricole et industrielle. Les barrières étanches de argiles compactés (CCL) sont de bonne alternatives pour la construction de décharge d'ordures au Brésil, parce que quelques régions ont des sédiments argileux et des argiles. L'état du São Paulo a un développement industriel important et 36 million de habitants.

Les formations rocheuses de le Bassin du Paraná qui constituent l'importante ressource de l'eau souterraine dénommé Aquifère Guaraní affleure sur une grande region de l'état du São Paulo. Le Bassin du Paraná est composée, dans l'état du São Paulo, par sédiments, roches sédimentaires et roches extrusives du Devonian au Crétacé. Ces roches sont recouvertes par les sédiments Cenozoique et il a des matières premières différentes pour utiliser comme barrières étanches de argiles compactés.

Quelque sols de la Formation Corumbataí utilisés dans cette étude ont été sélectionnés par la courbe granulométrique et minéralogie de l'argile. Les propriétés physiques des sols ont été déterminées par les essais caractérisation. Les minéraux argileux ont été déterminés par analyse thermique différentielle et diffractométrie des rayons X. La caractérisation physico-chimique a été utilisée pour avalier le compétence pour être utilisée comme barrière étanche. Les essais d'équilibre en série avec des métaux lourds ont été effectuée pour déterminer les isothermes de l'adsorption.

Les résultats de la analyse physico-chimique, de la composition minéralogique et des isothermes de l'adsorption indiquent que les materaux de la Formations Corumbataí peuvent être utilisés comme CCL.

Keywords: compacted clay liner, clayey soil, assessment properties

INTRODUCTION

Domestic and industrial waste increased during the last century because urban areas, industrial production and population also expanded. Waste disposal is of concern because of its potential adverse affect on the environmental. Drinking water supplies have to be protected from organic and inorganic leachate generated in municipal and industrial landfills. In order to develop adequate landfill design it is necessary evaluate the physical behaviour and attenuation characteristics of soils. An understanding of the transport mechanisms through liners is also important in order to prevent environment contamination (Rowe & Booker 1985).

In Brazil, São Paulo state is highly developed industrially with a large population that generates thousands tonnes of waste every day. This waste has been disposed of in adequate places to preserve the environment, as well as surface and groundwater. The Guaraní Aquifer is composed of the Botucatú (Triassic-Jurassic sandstones) and Serra Geral (Jurassic-Cretacic basalt) formations. It is the biggest groundwater resource in São Paulo state and supplies many cities in the middle-west of the state (Figure 1). It is necessary to choose adequate sites and material to build sanitary landfills to preserve the quality of the water resources of this region.

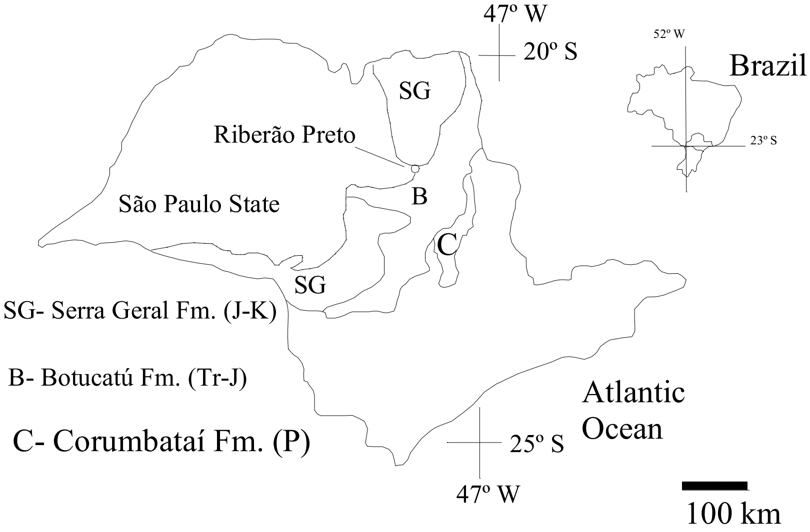


Figure 1 Extent of the Corumbataí, Botucatu and Serra Geral Formations in São Paulo state, southeastern Brazil.

Some clayey soils have the potential to use as raw material in sanitary landfill liners, as that of the Corumbataí Formation (Permian) in the middle-west of the state. This geological unit has been used as raw material by the ceramic industry, and it has layers with mineralogical and granulometric differences (Mason et al. 2000, Christofoletti et al. 2001). In some layers, the clay mineralogy is composed of different percentage of smectite, illite and kaolinite, and in other layers the clay mineralogy is composed of illite and kaolinite. The Corumbataí Formation has the potential for use as compacted clay liners (CCL). Therefore the objective of this paper is evaluate the potential use of the Corumbataí Formation for compacted clay liners. Accordingly its Mineralogical, physical and chemical properties were examined.

BACKGROUND

Sanitary landfill is placed on natural clayey soil or on a compacted clayey liner. There are differences in clay liner specifications (Yong et al 1999). Some clay liner specifications and properties cited by Yong et al (1993) are low hydraulic conductivity (< 10^{-9} m/s), minimum 10% clay content and plasticity index (PI) between 12% and 65%.

Some years ago the design of CCL was based on controlled leachate migration using low hydraulic conductivity of soils. If the solute migration through a liner is very slow, then it is necessary evaluate its long-term behaviour (Rowe & Booker 1985). The solute migration is controlled by different mechanisms such as advection, diffusion, sorption, and precipitation (Rowe & Booker 1985, Schackelford & Daniel 1991). If diffusion is the most important control mechanism of solute migration in fine soils, then it is necessary evaluate the chemical migration in such soils (Gooddall & Quigley 1977, Rowe & Booker 1985, Crooks & Quigley 1984, Schackelford & Daniel 1991).

It is necessary determine the physical properties of fine soils such as the specific gravity (Gs), liquid limit (LL) and plasticity index (PI), granulometric size, maximum dry density (γ_d),and hydraulic conductivity (k). Yong et al. (1999) suggested that complementary tests should be conducted such as the exchange cations capacity (CEC), specific surface area (SSA), clay mineralogy, pH, and electrical conductivity. If the soil has acceptable values in these tests then the batch equilibrium test should be conducted.

Research was conducted in the Department of Geotechnics (Engineering School São Carlos of São Paulo University) using mixed lateric soils from the Botucatú and Serra Geral formations. The target was to develop efficient and low cost technologies for CCL adapted to local soils (Paraguassú et al. 2002, Leite et al. 2003). The batch equilibrium test and column transport test were conducted involving different elements (K^{+1}, Cd^{+2}, Cu^{+2}, Cl^{-1}, F^{-1}). Kaolinite is the clay mineral present in these lateritic soils, but Fe, Al and Ti oxides and hydroxides are common. The hydraulic conductivity is below 10^{-9} m/s and physic-chemical properties are given in the Table 1. These lateritic soil mixtures were evaluated as suitable to use as CCL.

MATERIALS AND METHODS

The physical properties of the soil samples were determined using the normal simple tests specified in geotechnical investigations. The natural water content, specific gravity (Gs), liquid limit (LL), plasticity index (PI), and granulometric size were conducted in accordance with the Brazilian Standards NBR 6457, 6459, 6508, 7180, 7181. **The c**ation exchange capacity (CEC) and methylene blue value (VB) were determined using the methylene blue technique proposed by Lan (1977) and modified by Pejon (1992). The clay mineralogy was identified using differential thermal analysis in accordance with Grim (1953) and Mackenzie (1957). The **f**ree swell was determined from static compaction remolded samples in oedometers with Proctor parameters, and the test was conduced in accordance with Madsen (1999). At the same time the hydraulic conductivity was derived from the oedometer tests.

Table 1 Botucatu and Serra Geral soil mixture: hydraulic and physic-chemistry properties (Adsorption parameters by K^{+1} solution)

	CEC (cmol/kg)	**k (m/s)**	**K_d**	**Langmuir II b**	**Langmuir NL b ($cm^3/\mu g$)**	**Langmuir Sm (μg/g)**	**R_d**
Leite et al (2004)	2.2	Nd	Nd	0.015	1.12×10^{-2} a 4.4×10^{-3}	696 a 882	Nd
Paraguassu (2002)	2.2	1×10^{-8} a 8×10^{-10}	0.73-1.34	Nd	Nd	Nd	5 - 11

Batch equilibrium tests with KCl solution were conducted as indicated Roy et al. (1992) in order to determine sorption behaviour. The soil-solution ratio (SSR), equilibrium time and sorption isotherm were determined by the batch test. **F**lame atomic absorption spectrophotometry was used to obtain K^+ concentrations.

SSR values of 1:4, 1:10, 1:20, 1:40, 1:60, 1:100, 1:200, 1:500 were examined with an initial concentration of K^+ solution of 391 ppm (0.01M). The soil solution suspension was placed in 0.2 l Erlenmeyer covered with synthetic rubber plugs and mixed in a rotary shaker for 24 hs as recommended by Leite et al (2003). Suspensions were centrifuged at 3000 rpm in glass tubes for 15 min. After phase separation, the electrical conductivity and pH of the supernatant solution were recorded. All tests were performed at 22 ± 2ºC.

The adsorbed concentration was determined using Eq. (1) and used to choose the soil-solution ratio. The equilibrium test was performed on 10 samples for periods of 0.75 to 72 hs. Then the sorption isotherm was determined with initial concentrations of K^+ 0.1 M; 0.075 M; 0.05 M; 0.025 M; 0.013 M; 0.01 M; 0.0075 M; 0.005 M; 0.0025 M; 0.001 M.

$$S = \frac{(C_0 - C_e)V}{M} \tag{1}$$

S = mass of sorbed contaminant per unit mass of soil (μg/g), V = solution volume (l), M = dry mass of soil (g), C_0 = initial concentration and C_e equilibrium concentration (mg/l).

RESULTS AND DISCUSSION

Two soils were studied, soil (1) is from the Corumbataí Formation and soil 2 is a weathered soil from the same formation. Both soils are fine-grained but they have different physical properties. (Table 2). Soil 1 is a clayey silt soil and soil 2 is a silty clay soil. The physical properties values are considered suitable for use as CCL (Yong et al 1999).

Table 2 Physical soils properties

Soil	**Granulometric**			**γ_s (kN/m^3)**	**LL (%)**	**PI (%)**	**SUCS**	**γd (kN/m^3)**	**ω_{opt} (%)**
	Sand (%)	Silt (%)	Clay (%)						
1	21	56	23	2.69	38	14	CL	1.80	16.0
2	12	32	56	2.69	74	32	MH	1.54	24.2

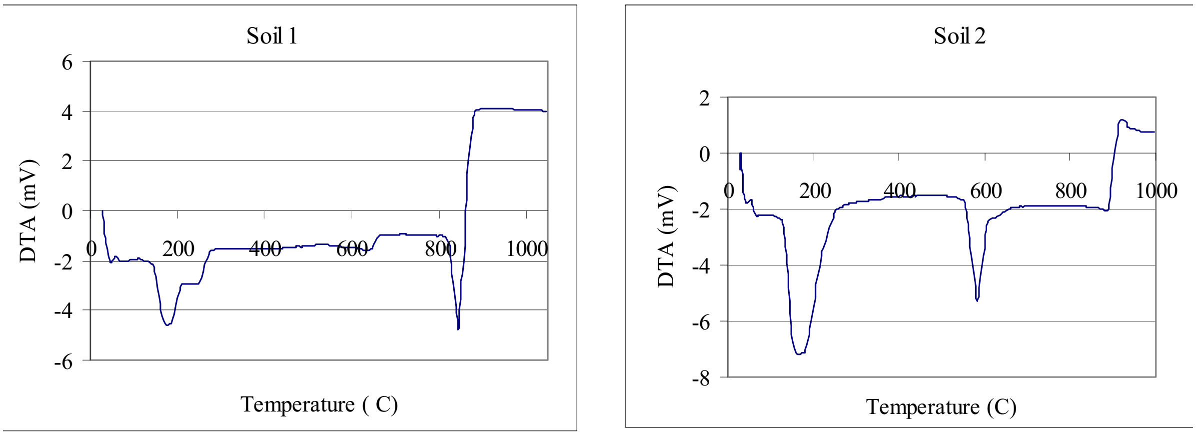
The physico-chemical soil properties are different. They have different CECs and specific surface areas and soil 1 has alkaline pH (Table 3). Calcite was determined in soil 1 by reaction with HCl.

The clay fraction of soil 1 has 52 cmol/kg of CEC and the clay fraction of soil 1 has 38 cmol/kg of CEC. Differential thermal analysis of soil 1 shows an endothermic double peak at 180 to 240ºC, a little endothermic peak at 660 ºC and an endothermic peak at 830ºC (Figure 2a). These peaks correspond to vermiculite-biotite intermixed or saponite-chlorite intermixed. The endothermic peak of 830°C also corresponded to calcite, and this could mask intermixed saponite-chlorite. The DTA of soil 2 shows endothermic peaks at 180ºC and 660ºC, and an exothermic peak in 950ºC. These peaks correspond with kaolinite and illite mixtures, and a small amount of smectite also could be present (Figure 2 b).

Table 3 Physico-chemical and mineralogical propiçertie

Soil	**CEC (cmol/kg)**	**VB**	**Specific surface (m^2/g)**	**pH H_2O**	**pH KCl**	**ΔpH**
1	12	3.9	96	9.03	5.50	3.53
2	21	6.7	165	7.62	3.75	3.87

The soils studied in the present work have marked differences with those studied by Paraguassú et al (1992) and Leite et al. (2003). The Corumbatai formation soils have more clay content, and the CEC values are 5 to 10 larger. The clay mineralogy is a mixture 1:1 and 2:1 clays. The oxides and hydroxides of Fe, Al and Ti were not identified and calcite was determined by DTA and the high pH in water.



a) b)

Figure 2. a) Soil 1 DTA. b) Soil 2 DTA

The free swell values are 1.8 % and 6.0 % for soil 1 and 2 respectively. The hydraulic conductivity value, using by destilled water, is 2 x10^{-10} m/s, and it is not significantly modified by the free swell.

An 1:20 SSR was chosen (Figure 3) and for this SSR, concentration solutions of 0.0075 M and 0.0132 M saturated the exchange capacity of soil 1 and soil 2 respectively.

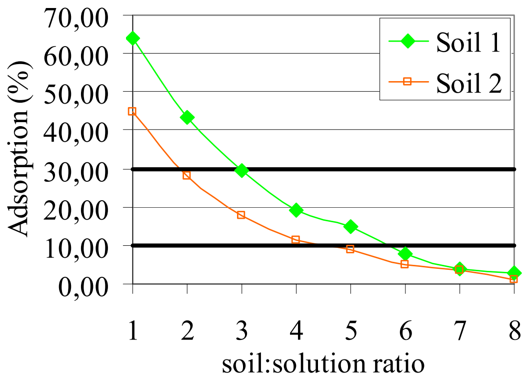
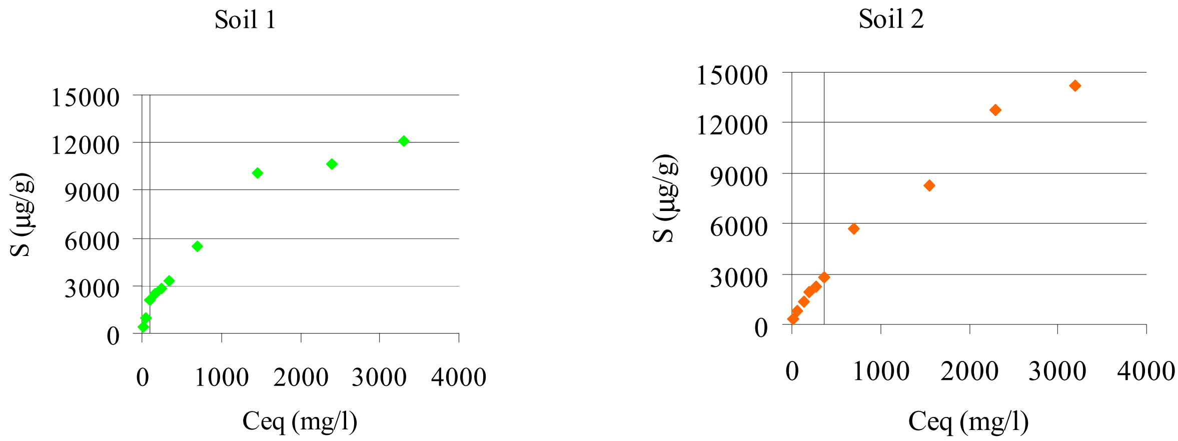


Figure 3 Soil: solution ratio determination

Sorption isotherms show other mechanisms besides cations exchange by clay minerals, because the soils “adsorbed” a much larger amount of K^+ than their CEC. Researches should be carried out to understand these mechanisms and how they could modify CCL properties.



a) b)

Figure 4 Sorption isotherms a) soil 1. b) soil 2

The Freundlich isotherm equation (Eq. 2) was used to predict the relation between the adsorbed and the equilibrium concentration,

$$S = K_f C^n \qquad (2)$$

S = mass of sorbed contaminant per unit mass of soil (µg/g), C = equilibrium concentration (mg/l), and K_f and n are the parameters adjusted to fit the experimental data. The values of Freundlich parameters are shown in Table 4 and good fits were obtained for the experimental values (R^2 greater than 0,97). Differences in the Freundlich parameters between soils were expected due to differences CEC values.

Table 4 Freundlich parameters .

Soil	**K_f**	**n**	**R^2**
1	83.34	0.6371	0.9714
2	37.45	0.7757	0.9962

CONCLUSIONS

The results obtained from geotechnical physical characterization tests on Corumbataí soils indicate that they attain the criteria set of Yong(1999) for CCL. These soils have higher CEC than soils mixtures of the Botucatú and Serra Geral Formations studied by Paraguasú et al. (2002) and Leite et al. (2003). Tlay mineralogy is different and the Corumbataí soils do not have Fe, Al and Ti oxides and hydroxides. Sorption isotherms show other mechanisms besides cations exchange of clay minerals. More batch tests and column tests must be carried out to understand these mechanisms and how they could modify CCL properties when used in a sanitary landfill liner. The physical characterization, physico-chemical and mineralogical values provide a good indication of the potential of Corumbataí soils for contaminant attenuation in CCL.

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