On the Compatibility and Theoretical Equations for Mixtures of Tropical Soils and Bentonite for Barrier Purposes

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Abstract. Vast regions of the globe are covered by tropical soils, increasing the perspective of using them for barrier design at solid/liquid disposal facilities. They comprise diverse pedological taxonomies, but have common features, specially the dominant mineral composition of quartz, kaolinite clay and Fe-Al-Mn oxides/hydroxides. Despite the vast occurrence, sometimes tropical soil samples may not be suitable for barrier purposes, which can be overcome using bentonite as additive, provide that technical evaluations are adopted. An important technical issue to be addressed is the compatibility between the barrier materials and the liquids to be disposed. Compatibility studies usually involve laboratory testing using direct and indirect methods, which were involved in this research. The indirect studies involved modified Atterberg limits and the direct approach was developed using direct permeation. The tropical soil samples were selected to be in different weathering stages, considering the laterization degree. The chosen bentonite content to compose the mixtures was 3, 6, 9 and 12% on a dry weight basis. The elected chemical solutions for these studies were: calcium chloride (CaCl₂), nitric acid (HNO₃), sodium hydroxide (NaOH), ethanol (C₂H₅OH) and tap water. As a summary, the main research objectives were: (1) comparison between direct and indirect methods for compatibility evaluation; (2) assessment of the influence of the bentonite content on the compatibility of tropical soil samples; (3) investigation of the influence of the laterization degree on compatibility and (4) evaluation of theoretical equations to predict the compatibility of these samples through back analysis. The results indicate that: indirect and direct methods are similar from a qualitative point of view; bentonite is more sensitive to chemicals than the natural soil components; the laterization degree is not so influent on the compatibility and good compatibility predictions were obtained with the proposed theoretical equations.

Keywords: bentonite, clayey barriers, compatibility predictions, modified Atterberg limits, theoretical equations, tropical soils.

1. Introduction

Tropical soils cover approximately 2.5 billion hectares on the globe (IBGE, 2005), a number that alone demonstrates the great potential of these materials for civil construction. They result from intensive weathering and pedological processes, mainly laterization, driven by the hot and humid climate of the tropics (Van Breemen & Buurman, 2003). Laterization leads to peculiar mineralogy, mainly composed of quartz, kaolinite and Fe-Al-Mn oxides/hydroxides, standing on a very loose and porous structure.

The similarity in terms of mineralogical composition of tropical soil samples is not reflected by their variety of pedological classifications, as showed by the taxonomies presented on Table 1. Since these terms are not familiar to the geotechnical community, precious information is often neglected in the engineering practice.

Nogami & Vilibor (1995) have drawn the attention of the geotechnical community to the potentiality of these materials for civil construction. They result from intensive weathering and pedological processes, mainly laterization, driven by the hot and humid climate of the tropics (Van Breemen & Buurman, 2003). Laterization leads to peculiar mineralogy, mainly composed of quartz, kaolinite and Fe-Al-Mn oxides/hydroxides, standing on a very loose and porous structure.

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This system is based on the results of the laboratory compaction test Mini-MCV (Moisture Condition Value) and the weight loss by immersion in water. The main soil orders of the MCT system refers to sandy, clayey, lateritic and non-lateritic soil specimens. Usually, the lateritic character is favorable for compaction. Later, Vertamatti (1998) introduced a transitional order between the lateritic and non-lateritic soils, calling the new system MCT-M, which will be used later in this study.

Sometimes, however, the decrease in hydraulic conductivity ($k$) induced by compaction may not be sufficient for barriers, as pointed out by Lukiantchuki & Esquivel (2010). In these cases, the use of bentonite as admixture is interesting, considering the well-known ability of this clay to reduce hydraulic conductivity and increase sorption (e.g. Ebina et al., 2004; Akbulut & Saglam, 2004; Osinubi & Nwaifu, 2006; Osinubi et al., 2009; Morandini & Leite, 2013; Akcanca & Aytekin, 2014; Morandini & Leite, 2015a).
Another important issue for barrier design is the capacity of the compacted clay liner (CCL) to maintain its properties after contacting with the different contaminant fluids to be disposed. For instance, these contaminant solutions may involve municipal solid waste leachate, urban sewage, effluents from industrial and mining activities and others. This capacity is called “liner compatibility”, which is very important for the long term performance of the barrier.

It is clear that compatibility depends upon complex soil-fluid interactions, which are not easy to identify and understand. So, most of the methods for investigating compatibility aim to recognize the changes in the mechanical properties of the soil specimens, instead of identifying specific “solid-fluid” reactions that take place mainly at the microscopic scale.

Laboratory works on compatibility are often classified as “indirect” and “direct” methods (Shackelford, 1994). The indirect methods investigate the changes in soil index properties, such as Atterberg Limits, for instance, after contact with the aggressive fluids that will be disposed. It only gives a perspective of the soil compatibility, once the “direct properties” of the soil that are involved in the barrier performance are not directly evaluated, such as hydraulic conductivity, swelling potential, shear strength and others.

Examples of the application of indirect methods for clays (bentonite and kaolinite) and tropical soils, using different chemical solutions (organic, saline, acid and alkaline), can be found on Bowders & Daniel (1987), Budhu et al. (1990), Egloffstein (2001), Farnezi & Leite (2007), Bouazza et al. (2007) and Katsumi et al. (2008). These authors have used modified Atterberg limits and/or free swelling tests for evaluating compatibility, finding a reduction in the plasticity as well as in the swelling potential of the clays, particularly when saline solutions and concentrated organic solutions with low dielectric constant were percolated. They have also concluded that the liquid limit was more sensitive than the plastic limit, and divalent ions had preference over monovalent ions. Most of these conclusions were consistent with the expected effect of the reduction in the double-layer thickness of clay particles, as pointed out by Mitchell (1993).

Direct studies, in turn, evaluate the changes induced by the contaminant liquids in properties that will be directly involved in the barrier performance. They certainly provide a more realistic scenario of what the barrier will face in the field, as pointed out by Budhu et al. (1990), Shackelford (1994), Shackelford et al. (2000) & Kinsela et al. (2010). Nevertheless, percolating soil with chemical solutions is not always feasible, since the laboratory testing apparatus must resist corrosion and other possible damage caused by the chemicals from the contaminant solutions. As most of the ordinary equipment found in conventional geotechnical laboratories may not support these solutions, direct percolation sometimes is restricted.

Interesting reports on the direct percolation of chemical solutions on clayey soil samples can be found in Stewart et al. (2003), Sunil et al. (2006 & 2009), Nayak et al. (2007), Katsumi et al. (2008) and Kinsela et al. (2010). Those papers have demonstrated that organic and salty solutions, as well as actual and synthetic leachates have reduced the plasticity and increased the hydraulic conductivity of the studied soil samples.

Morandini & Leite (2015a,b) describe the modifications imposed to tropical soils, a result of the bentonite addition, in terms of hydraulic conductivity and shear strength, respectively. The studied soil samples were lateritic, transitional and non-lateritic specimens of tropical soils according to the MCT-M system (Vertamatti, 1998), using bentonite proportions of 0, 3, 6, 9 and 12% (dry weight basis) to compose mixtures. Laboratory works have included: 1) mineralogical, geotechnical and chemical characterization; 2) estimation of hydraulic conductivity (flexible-wall permeameter); 3) shear strength and 4) direct and indirect compatibility. The findings of the items 1 through 3 have been described by Morandini & Leite (2015a,b), while compatibility (item 4) is treated in the present paper.

So, direct and indirect compatibility studies were performed using the following chemical solutions: calcium chloride (CaCl2), nitric acid (HNO3), sodium hydroxide (NaOH), ethanol (C2H5OH) and tap water. As a summary, the research objectives of this specific study were: (1) comparison between direct and indirect methods for compatibility evaluation; (2) assessment of the influence of the bentonite content on the compatibility of tropical soil sam-

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**Table 1 - Pedologic orders of tropical soils according to different taxonomies (Source: Morandini & Leite, 2015a).**

| Brazilian Soil Classification System | WRB/FAO | U.S. Taxonomy |
|-------------------------------------|---------|---------------|
| Latosols                            | Ferralsols | Oxisols       |
| Clay soils                          | Lixisols | Ultisols     |
| Acrisols                            | Alisols | Oxisols / Ultisols |
| Nitisols                            | Nitisols | Oxisols / Ultisols |
| Plintosols                          | Plinthosols | Oxisols       |

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amples; (3) investigation of the influence of the laterization degree on the compatibility and (4) evaluation of theoretical equations to predict the compatibility of these samples through back analysis.

2. Theoretical Propositions

As mentioned before, in spite of the variety of the pedological orders of the tropical soils showed on Table 1, their mineralogical composition is dominated by quartz, kaolinite and Fe-Al-Mn oxides/hydroxides. The concentration of oxides/hydroxides can be part of the parental rock heritage, but it is also related to the stage of the weathering process (mainly hydrolysis) and pedological evolution.

In terms of electrical charges, the clay fraction of tropical soils is a mix of permanent charged minerals mainly provided by the kaolinite clay, with variable charged solids, such as Fe-Al-Mn oxide/hydroxide (Yong et al., 1992 and Yu, 1997). Most of the permanent charges of the kaolinite are negative and result from isomorphic substitution and lattice breaking at the mineral genesis (Yong et al., 1992). The variable charges, in turn, are related to the specific sorption of the radicals H⁺ and OH⁻ from the water molecule breaking, mainly controlled by the pH conditions of the soil solution (Grim, 1968 and Yu, 1997).

The constant charge density (σₖ) at the midpoint distance between two clay particles on the clay-electrolyte system can be modeled by Eq. 1 (Mitchell, 1993), where cₒ is the ionic concentration (ions/m³), zᵣ the ionic valence, e the electric charge (1.602 x 10⁻¹⁹ Coulomb), ψ the electric potential, k the Boltzmann constant (1.38 x 10⁻²³ JK⁻¹) and T the absolute temperature (in Kelvin). Equation 1 is usually referred as the Guy-Chapman model (Mitchell, 1993) and can be used also on the estimation of variable charge density of variable charge soils (σᵥ), but in this case the electric potential (ψ) is constant (Yu, 1997).

As pointed out by Mitchell (1993), two equations were used to compose Eq. 1: (1) the potential function (ψᵥ), expressed by Eq. 2, being x a dimensional quantity equivalent to 1 Å, e the electronic charge, and (2) (1/λ) the distance function which is related to the electric permittivity (ε), as can be seen in Eq. 3.

\[ \sigma_v = \sqrt{8c_{\text{Cl}}ekT} \sinh \frac{ze^\psi}{2kT} \]  

\[ \psi_v = \frac{ze^\psi}{kT} \]  

\[ \frac{1}{\lambda^2} = \frac{2c_{\text{Cl}}z_i^2e^2}{ekT} \]  

The quantity λ expresses the physical space of the gravity centroid between two charged particles and represents the ionic double layer thickness, which can be calculated using Eq. 4, where \( e_0 \) is the vacuum permittivity (8.85 x 10⁻¹² C²J⁻¹m⁻¹) and D is the dielectric constant of the pore solution.

\[ \lambda = \sqrt{\frac{e_0DKT}{2e_{\text{Cl}}z_i^2e^2}} \]  

Equations 1 through 4 show that the compatibility of clay can be influenced by the differences in the chemical composition of the pore solution, since the ionic double layer thickness and the plasticity and hydraulic properties of the soils are closely related.

The ionic double layer thickness is estimated using the ionic strength (I = 0.5Σcᵢᵢzᵢ²) and the dielectric constant (D) of the pore solution for constant temperature conditions (Bouazza et al., 2007 and Katsumi et al., 2008). Therefore, a theoretical compatibility index (Rₑb) was formulated to estimate the compatibility of pure bentonite samples by the correlation of the water ionic strength (Iₚ) and dielectric constant (Dₑ) to the pore solution parameters (Iₑ and Dₑ), as presented in Eq. 5. When the Rₑb is one, full compatibility exists of the pore solution relative to water, while Rₑb values smaller or greater than one indicate incompatibility.

\[ R_{\text{eb}} = \sqrt{\frac{I_{\text{b}}D_{\text{p}}}{I_{\text{eb}}D_{\text{eb}}}} \]  

As suggested by Bolt (1955), \( I_b = 0.25 \) M for pure sodium bentonite clay; in turn, \( I_{\text{eb}} \) can be estimated by the geometric mean between the ionic strength of the water \( I_p \) and pore solution \( I_b \), multiplied by the cation activity (α), as presented in Eq. 6 (Langmuir, 1997).

\[ I_{\text{eb}} = \sqrt[3]{(\alpha I_p) \lambda} \]  

As mentioned before, calcium chloride (CaCl₂) 1 M and ethanol (C₂H₅OH) solutions were used in the indirect and direct compatibility tests of this research, so the \( R_{\text{eb}} \) values for these solutions were estimated as follows.

Considering the CaCl₂ 1 M solution: \( I_b = 2.0 \) M; \( \alpha_{\text{Ca}} = 0.293; \alpha_{\text{Na}} = 0.734 \), which leads to \( I_{\text{eb}} = 0.607 \) and \( R_{\text{eb}} = 0.64 \). For the ethanol solution, the dielectric constant \( D_b \) is 24.3, resulting in \( R_{\text{eb}} = 0.55 \). In these cases, the theoretical compatibility for both solutions is smaller than one, which indicates loss of plasticity, being the ethanol solution more “incompatible” to the bentonite sample than the CaCl₂ 1 M solution.

Since the premises adopted for calculating the \( R_{\text{eb}} \) by Eq. 5 cannot be directly applied to the mixtures (bentonite + tropical soil), an expedite model \( (R_{eb}) \) was formulated using the proportion of bentonite clay (%B) of the mixture and also its natural clay content, which is almost exclusively composed of kaolinite. As this last clay presents an interlayer expansion length in the order of 7 to 12 Å, while for the bentonite it is 80 Å, it is reasonable to think that the effect of any pore solution to the kaolinite clay is only 15%
(12 Å/80 Å) compared to the bentonite, as presented by Eq. 7.

3. Materials and Methods

3.1. Summary of the sample properties

The bentonite clay used in the tests is commercially available in Brazil with the denomination Brasgel PA®. The soil samples of lateritic, transitional and non-lateritic behavior, according to the abacus of Vertamatti (1998), were collected in the vicinities of Ouro Preto, state of Minas Gerais, Brazil.

Mixtures using these soils and the commercial bentonite were manually composed. The sampling locations and preparation, as well as the mixing and characterization methods and results are fully reported by Morandini & Leite (2015a). Table 2 presents only the sample designations, including the bentonite proportions of the mixtures (dry weight basis), and Table 3 reprints a summary of the geotechnical and physicochemical properties as presented by Morandini & Leite (2015a).

3.2. Indirect compatibility tests

Modified plastic ($\omega_p$) and liquid ($\omega_l$) limits were determined in the laboratory using the solutions and concentrations described next, as they represent the classes of organic and inorganic liquids miscible in water presented by Shackelford (1994). The test procedures followed the Brazilian standards NBR 6459 (ABNT, 2016) for the latter and NBR 7180 (ABNT, 2016) for the former. Special care was taken for handling these solutions because of their aggressive nature.

- Water: tap water
- Salt: CaCl$_2$ at the concentration of 1 M (ionic strength = 2 M = 40.1 mg/L),
- Organic: Ethanol (C$_2$H$_5$OH), PA (dielectric constant = 24.3)
- Acid: HNO$_3$ diluted to pH = 3
- Base: NaOH diluted to pH = 11

Besides measuring $k$ in the laboratory with different permeation fluids, this parameter was also estimated theoretically ($k_M$) through the proposition of Morandini & Leite (2015a) shown in Eq. 10. It is a variation of the well-known Kozeny-Carman ($KC$) equation that includes the mesoplastic point (MP), the $C_{KC}$ coefficient (a shape factor related to the geometry of the soil pores); $\gamma_f/\mu$, respectively for the unit weight and dynamic viscosity of the percolating fluid; $e$ the soil void ratio and $a$ and $b$, the linearization parameters for these soil samples as referred in Morandini & Leite (2015a).

$$k_M = \frac{C_{KC}}{\gamma_f} \frac{e^3}{\mu (1 + e)} aM^b$$

(10)

$$k_M = \frac{C_{KC}}{\gamma_f} \frac{e^3}{\mu (1 + e)} 8.32 \times 10^{-4} M^{-4.48}$$

(11)

$$k_M = \frac{C_{KC}}{\gamma_f} \frac{e^3}{\mu (1 + e)} 2.95 \times 10^2 M^{-7.56}$$

(12)

$$k_M = \frac{C_{KC}}{\gamma_f} \frac{e^3}{\mu (1 + e)} 4.68 \times 10^1 M^{-7.26}$$

(13)

3.3. Direct compatibility tests

3.3.1. Sample preparation

Compacted soil specimens of the mixtures were prepared under Normal Proctor energy at 2% wet of optimum moisture content using the tested chemical solutions and tap water. These compaction effort and moisture content are ordinary for landfill liner construction in Brazil and many other countries. The specimens were 5 cm in diameter and 10 cm high. Their bentonite content (dry weight basis) was chosen according to the criterion that they must show a minimum $k$-value of $5 \times 10^{-8}$ cm/s under an effective confining stress of 40 kPa. This $k$-value meets most of the international standards for barrier design (Koch, 2002), and the confining stress of 40 kPa is ordinary for municipal solid waste landfills. So, according to Fig. 1, the proportions that meet these criterion on a dry weight basis, called here as “optimum bentonite content”, were: SL$_{ot}$ = 9%; ST$_{ot}$ = 4% and SN$_{ot}$ = 2%.

Table 2 - Sample designation and bentonite proportions (dry weight basis).

| Lateritic samples | Transitional samples | Non-lateritic samples |
|-------------------|----------------------|-----------------------|
| Designation       | Bentonite (%)        | Designation           | Bentonite (%)        | Designation       | Bentonite (%)        |
| SL                | 0.0                  | ST                    | 0.0                  | SN                | 0.0                  |
| SL03              | 3.0                  | ST03                  | 3.0                  | SN03              | 3.0                  |
| SL06              | 6.0                  | ST06                  | 6.0                  | SN06              | 6.0                  |
| SL09              | 9.0                  | ST09                  | 9.0                  | SN09              | 9.0                  |
| SL12              | 12.0                 | ST12                  | 12.0                 | SN12              | 12.0                 |
Table 3 - Summary of the geotechnical and physicochemical properties of the samples (Reprinted from Morandini & Leite, 2015a).

| Property                        | Sample | SL | SL03 | SL06 | SL09 | SL12 | ST | ST03 | ST06 | ST09 | ST12 | SN | SN03 | SN06 | SN09 | SN12 | BENT |
|---------------------------------|--------|----|------|------|------|------|----|------|------|------|------|----|------|------|------|------|------|
| Liquid limit $\omega_L$ (%)     |        | 23 | 42   | 57   | 72   | 87   | 52 | 78   | 96   | 117  | 156  | 56 | 76   | 96   | 117  | 136  | 683  |
| Plastic limit $\omega_p$ (%)    |        | 12 | 21   | 23   | 26   | 27   | 30 | 28   | 29   | 27   | 28   | 31 | 34   | 37   | 40   | 44   | 91   |
| Plasticity index $PI$ (%)       |        | 11 | 21   | 33   | 47   | 60   | 22 | 50   | 67   | 90   | 128  | 25 | 42   | 59   | 76   | 92   | 592  |
| Mesoplastic point $MP$ (%)      |        | 17 | 32   | 40   | 49   | 57   | 41 | 53   | 62   | 72   | 92   | 44 | 55   | 67   | 79   | 90   | 387  |
| Density of solids $\rho_s$ (g/cm$^3$) |    | 2.90 | 2.89 | 2.88 | 2.86 | 2.84 | 2.84 | 2.83 | 2.82 | 2.80 | 2.79 | 2.76 | 2.75 | 2.74 | 2.73 | 2.72 | 2.45 |
| Grain size (%)                  |        |    |      |      |      |      |    |      |      |      |      |    |      |      |      |      |      |
| Clay                            |        | 29 | 29   | 30   | 30   | 31   | 41 | 39   | 41   | 42   | 36   | 38 | 42   | 44   | 46   | 91   |      |
| Silt                            |        | 16 | 18   | 17   | 20   | 17   | 9  | 10   | 9    | 9    | 10   | 11 | 11   | 11   | 12   | 9    |      |
| Fine sand                       |        | 35 | 34   | 34   | 29   | 28   | 36 | 17   | 17   | 18   | 19   | 23 | 20   | 20   | 19   | 20   | 0    |
| Medium sand                     |        | 16 | 15   | 15   | 15   | 17   | 8  | 27   | 26   | 25   | 24   | 18 | 19   | 19   | 17   | 14   | 0    |
| Coarse sand                     |        | 3  | 3    | 4    | 5    | 7    | 5  | 6    | 6    | 6    | 6    | 10 | 9    | 6    | 6    | 6    | 0    |
| Gravel                          |        | 1  | 1    | 0    | 1    | 0    | 1  | 1    | 1    | 1    | 1    | 3  | 3    | 3    | 3    | 2    | 0    |
| USCS classification             |        | CL | CL   | CH   | CH   | CH   | CH | CH   | CH   | CH   | MH   | CH | CH   | CH   | CH   | CH   | CH   |      |
| Activity (Skempton)             |        | 0.38 | 0.71 | 1.11 | 1.55 | 1.94 | 0.54 | 1.28 | 1.63 | 2.19 | 3.05 | 0.68 | 1.10 | 1.39 | 1.73 | 2.01 | 6.50 |
| Normal proctor compaction       |        |    |      |      |      |      |    |      |      |      |      |    |      |      |      |      |      |
| $\rho_{max}$ (g/cm$^3$)         |        | 1.82 | 1.81 | 1.79 | 1.78 | 1.76 | 1.62 | 1.66 | 1.62 | 1.60 | 1.59 | 1.62 | 1.61 | 1.60 | 1.59 | 1.58 |      |
| $w_a$ (%)                       |        | 12.5 | 14.5 | 16.2 | 17.8 | 19.1 | 21.5 | 22.5 | 23.3 | 24.0 | 24.8 | 22.2 | 23   | 23.6 | 24.4 | 25.1 |      |
| Saturation at $w_a$ (%)         |        | 61.0 | 70.5 | 77.0 | 83.8 | 88.6 | 88.4 | 89.8 | 88.8 | 89.7 | 91.4 | 87.1 | 89.2 | 90.3 | 92.5 | 94.3 |      |
| Cation exchange capacity (cmol/kg) | 3.6 | 6.2 | 9.8 | 12.9 | 15.7 | 6.9 | 13.5 | 18.6 | 25.3 | 36.0 | 7.9 | 11.6 | 16.0 | 20.3 | 24.8 | 99.4 |      |
| pH at 1:2.5 soil:solution $H_2O$ | 5.35 | 6.19 | 7.14 | 8.03 | 8.79 | 5.15 | 7.76 | 8.24 | 8.63 | 8.96 | 4.84 | 5.65 | 6.68 | 7.47 | 8.24 | 9.92 |
| pH at 1:2.5 soil:solution $KCl$  | 6.16 | 6.44 | 6.86 | 7.54 | 7.93 | 5.21 | 5.94 | 6.58 | 7.06 | 7.33 | 4.60 | 5.03 | 5.43 | 6.00 | 6.39 | 8.77 |
| $\Delta pH$                     |        | 0.81 | 0.25 | -0.28 | -0.49 | -0.86 | 0.06 | -1.82 | -1.66 | -1.57 | -1.63 | -0.24 | -0.62 | -1.25 | -1.47 | -1.85 | -1.15 |
| Electrical cond. (mS/cm)        |        | 0.09 | 0.28 | 0.5  | 0.67 | 0.84 | 0.84 | 0.43 | 0.68 | 0.87 | 0.97 | 0.08 | 0.44 | 0.78 | 1.02 | 1.35 | 1.3  |
| Specific Srface - $S_o$ (m$^2$/g) | 28.2 | 48.4 | 76.3 | 100.4 | 122.3 | 53.8 | 105.3 | 145.6 | 197.3 | 281.3 | 61.3 | 90.5 | 124.7 | 158.7 | 193.6 | 775.9 |

*NR - Not Rated.
3.3.2. Inert permeability cells

Flexible wall permeability tests under constant head conditions were applied to determine the $k_w$ and $k_f$ of the samples. Usually these cells are an adaptation from the conventional triaxial cells, that usually have many metallic parts vulnerable to corrosion when chemical solutions are percolated. To avoid corrosion, two “inert” triaxial cells were designed using only plastic materials, one for permeability determination and the other as a solution reservoir (Fig. 2). The parts of this new equipment are described next:

- Not welded walls made of acrylic (polymethyl-methacrylate);
- Base and top made of high density nylon;
- 0.5” low density polyethylene tubes;
- Stainless steel connections.

3.3.3. Test conditions

The schematic of the test apparatus used in soil percolation is depicted in Fig. 3. Pressure application systems were installed at the base ($p_1$) and over the top ($p_2$) of the soil specimens, along with the confining pressure system ($\sigma'$). A transducer for pore pressure ($u$) monitoring and an electronic gage for flow measurement were also coupled to the triaxial cell.

The test method followed the recommendations of Head (1986) and Morandini & Leite (2013), including backpressure saturation and pre-consolidation of the samples using the chemical solutions and tap water as permeation fluid. The backpressure saturation was limited to 300 kPa and full saturation was considered when the Skempton $B$ parameter reached a minimum of 0.94. In turn, pre-consolidation was induced by applying a total confining stress ($\sigma'$) of 323 kPa, leading to a constant effective confining stress ($\sigma''$) of 40 kPa, used for pre-consolidation was well as for percolation.

Once these procedures were terminated, percolation was imposed to the soil specimens by the application of a $p_1$ of 300 kPa, while $p_2$ was kept constant at 250 kPa. These conditions have produced a porepressure ($u$) of 283 kPa, a hydraulic head of 50 kPa and a hydraulic gradient ($i$) in the order of 50. So, $k$ values were determined using Darcy’s Law under constant head conditions.

Some aspects of these tests must be highlighted, as pointed out in Morandini & Leite (2013): (1) because of op-

Figure 1 - Bentonite content necessary to achieve a hydraulic conductivity of $5 \times 10^{-8}$ cm/s under a confining stress of 40 kPa, called “optimum bentonite content”: (a) SL$_{opt}$ sample; (b) ST$_{opt}$ sample and (c) SN$_{opt}$ sample (Morandini & Leite, 2015a).

Figure 2 - Inert triaxial chambers: (a) triaxial chamber and (b) solution reservoir.

Figure 3 - Schematic of the apparatus used in the percolation tests (Reprinted from Morandini & Leite, 2013).
erational reasons, including difficulties in soil compaction, moulding, saturation and consolidation, the hydraulic conductivity of the pure bentonite samples was not measured; (2) the rule \( \sigma_1 > p_1 > p_2 \) has been complied with to avoid soil liquefaction induced by excessive pore pressure increase; (3) the reported \( k_1 \) and \( k_2 \) values are the arithmetic average of 3 measures.

4. Results and Discussion

4.1. Indirect compatibility

Some of the bentonite properties are presented in Table 4, including the conventional and modified Atterberg limits, the mesoplastic point (MP), the indirect compatibility index (\( R_c \)) and the bentonite theoretical compatibility index (\( R_{cb} \)). As the pH variation induced by the acid and alkaline solutions was not incorporated into the compatibility model of Eq. 7, the \( R_{cb} \) was not calculated for the HNO\(_3\) and NaOH solutions.

It is clear from Table 4 that the ethanol (C\(_2\)H\(_5\)OH) and calcium chloride (CaCl\(_2\)) solutions had the major influence on the plasticity of the bentonite sample relative to the other solutions, as demonstrated by the variation of the \( R_c \) and \( R_{cb} \) values. It must be also pointed out the similarity between the theoretical compatibility index (\( R_{cb} \)) and the laboratory mesoplastic-point-based compatibility index (\( R_c \)) for these last solutions.

As can be seen in Fig. 4, the influence of the solutions on the bentonite properties was more accentuated for the liquid limit compared to the plastic limit, which has reflected also in the shape of the PI and MP curves. This result was consistent with the literature, as mentioned in the Introduction section.

Table 5 and Fig. 5 present the conventional and modified Atterberg limits as well as the \( R_c \) values for the tropical soils samples and their mixtures.

Figure 5 illustrates that the use of water as the molding fluid for the natural soils samples (SL, ST and SN) resulted in a small variation between the modified and conventional Atterberg limits, so it is concluded that the bentonite clay content was the main responsible for the increase in their incompatibility.

Figure 6 depicts the relation \( R_c \) vs. bentonite content of the soil samples and some excerpts of these diagrams are

| Properties                     | Chemical solutions |
|--------------------------------|--------------------|
|                                | Water     | CaCl\(_2\) | C\(_2\)H\(_5\)OH | HNO\(_3\) | NaOH |
| Liquid limit (\( \omega_L \)) (%) | 683       | 447       | 391              | 715      | 620  |
| Plastic limit (\( \omega_P \)) (%) | 91        | 63        | 52               | 103      | 83   |
| Plasticity Index (PI) (%)      | 592       | 384       | 340              | 612      | 537  |
| Mesoplastic Point (MP) (%)     | 387       | 255       | 221              | 409      | 352  |
| Indirect compatibility Index (\( R_c \)) | 1.00 | 0.66 | 0.57 | 1.06 | 0.91 |
| Theoretical compatibility Index (\( R_{cb} \)) | 1.00 | 0.64 | 0.55 | -     | -    |

| Solution | SL | SL03 | SL06 | SL09 | SL12 | ST | ST03 | ST06 | ST09 | ST12 | SN | SN03 | SN06 | SN09 | SN12 |
|----------|----|------|------|------|------|----|------|------|------|------|----|------|------|------|------|
| Water    | 23 | 42   | 57   | 72   | 87   | 52 | 78   | 96   | 117  | 156  | 56 | 76   | 96   | 117  | 136  |
| CaCl\(_2\) | 22 | 39   | 53   | 66   | 80   | 51 | 75   | 90   | 107  | 137  | 55 | 74   | 89   | 106  | 120  |
| C\(_2\)H\(_5\)OH | 21 | 38   | 52   | 65   | 77   | 50 | 74   | 89   | 105  | 134  | 54 | 73   | 88   | 104  | 118  |
| HNO\(_3\) | 22 | 42   | 56   | 74   | 88   | 52 | 79   | 96   | 118  | 158  | 56 | 75   | 95   | 108  | 138  |
| NaOH     | 22 | 41   | 55   | 69   | 83   | 51 | 76   | 94   | 113  | 150  | 55 | 75   | 93   | 114  | 130  |
worth mentioning: (1) the addition of bentonite has led to an increase in the incompatibility of the soil samples denoted by the linear decrease of the $R_c$ values, which corroborates the above analysis of Fig. 5; (2) a decrease in the $R_c$ value indicates loss of plasticity relative to water, which was the case presented by the ethanol, calcium chloride and sodium hydroxide solutions; (3) the acidic solution resulted in a slight increase in the plasticity for the SL and ST samples, while it was oscillatory for the SN mixtures; (4) of all natural soil samples (without any bentonite addition), the SL sample had the greatest incompatibility denoted by the lowest $R_c$ values along the ordinate axis; (5) in spite of the variability of the $R_c$ values of the natural soils (0% of bentonite), as shown in Fig. 6(a), the similarity among all diagrams indicates that the laterization stage had little influence on the general compatibility.

Satisfactory correlations between laboratory $R_c$ (Eq. 8) and theoretical $R_{cM}$ (Eq. 7) for ethanol and calcium chloride solutions can be observed in Fig. 7, since differences of no more than 4% between these two variables were found. It is also observed that the best fit was reached for the SL sample (Figs. 7a and 7b).

In general terms, the $R_c$ values for all soil samples and chemical solutions are in the range from 0.93 to 1.01, meaning more compatibility for these samples when compared with the pure bentonite clay values, whose values varied from 0.57 (C$_2$H$_5$OH), 0.66 (CaCl$_2$), 0.91 (NaOH) to 1.06 (HNO$_3$).

The influence of the weathering stage of the natural soil samples on the compatibility was analyzed by plotting the weathering index $K_i$ vs. $R_c$ (Fig. 8). $K_i$ is defined as $1.7 \times \frac{SiO_2}{Al_2O_3}$ (IBGE, 2005), so a small $K_i$ value refers to advanced weathering and vice versa. The $K_i = 2.0$ of the kaolinite clays is taken as a reference.

The $K_i$ index values for the samples SL, ST and SN are respectively 1.72, 2.36 and 3.08 (Morandini & Leite, 2015a), and no clear dependence of the $R_c$ on $K_i$ is observed since the curves for ST and SN are very similar.

### 4.2. Direct compatibility

The laboratory hydraulic conductivity ($k$) and the relative hydraulic conductivity measured with chemical solutions ($k_r$) for the samples compacted at the “optimum bentonite content” are presented on Table 6. It is clear from this table that the permeation with the other chemical solutions increased $k$ relative to water, as confirmed by their $k_r$ values greater than one. An exception is made for the lateritic soil (SL$_{opt}$ sample), since its permeation with HNO$_3$ slightly reduced $k$ relative to water ($k_r < 1 = 0.87$).

The $k_r$ values from Table 6 denote a significant increase (from 2.3 to 6.1) in $k$ when the ethanol solution was permeated. Daniel & Liljestrand (1984), Fernandez & Quigley (1985), Daniel et al. (1993) and Shackelford (1994)
also reported on the increase of $k$ for different clays due to contact and percolation with organic solutions of low dielectric constant, as is the case of the ethanol PA reagent (24.3) used in this research.

A simple comparison between $k_r$ values from Table 6 (direct compatibility) and $R_c$ values from Table 5 and Fig. 5 (indirect compatibility) reveals the same order of incompatibility for all the samples and solutions, as follows: $\text{C}_2\text{H}_5\text{OH} > \text{CaCl}_2 > \text{NaOH} > \text{HNO}_3$. This fact increases the perspective of using the modified Atterberg limits as a preliminary tool for evaluating the compatibility of soils for barrier purposes.

Table 7 summarizes the soil parameters used estimate $k_M$ values according to Eq. 11. A value $C_{KC} = 0.2$ was applied as suggested by Chapuis & Albertin (2003) when dealing with clayey soils. Table 8 presents the mesoplastic point...
(MP) values, the fluid parameters ($\gamma/\mu$) and the resultant $k_m$. These MP values were calculated from Eqs. 5 through 7 for the CaCl$_2$ and C$_2$H$_5$OH solutions; laboratory MP values were used for HNO$_3$ and NaOH solutions.

Figure 7 - Correlation between the $R_c$ and $R_{cm}$ values: (a) SL sample/CaCl$_2$; (b) SL sample/C$_2$H$_5$OH; (c) ST sample/CaCl$_2$; (d) ST sample/C$_2$H$_5$OH; (e) SN sample/CaCl$_2$ and (f) SN sample/C$_2$H$_5$OH.

Figure 8 - Weathering index $K_i$ vs. $R_c$ for all the natural soil samples and chemical solutions.
The correlation between the $k$ measured in the laboratory and the theoretical $k_M$ from Table 8 is depicted in Fig. 9(a) as function of the soil samples, and 9(b) as a function of the chemical solutions. Good fits were observed for all samples and solutions, exception made for the SN$_{ot}$ sample (pointed in Fig. 9a) and the C$_2$H$_5$OH solution (see Fig. 9b), whose measured $k$ is smaller than $k_M$, but remaining in the same order of magnitude.

If the theoretical propositions presented here are directly connected to the double layer theory in which bentonite clay is clearly inserted, it is reasonable to think that the $k_M$ values estimated for the mixtures with greater bentonite content should give better fit to the models, as was the case of the samples SL$_{ot}$ (9%) and ST$_{ot}$ (4%) compared to the sample SN$_{ot}$ (2%).

### Table 6 - Hydraulic conductivity ($k$) and relative hydraulic conductivity ($k_r$) for the “optimum bentonite content” samples with different solutions.

| Sample | Chemical solutions | Water ($k_r$) | CaCl$_2$ ($k_r$) | C$_2$H$_5$OH ($k_r$) | HNO$_3$ ($k_r$) | NaOH ($k_r$) |
|--------|-------------------|---------------|-----------------|----------------------|----------------|----------------|
| SL$_{ot}$ | 6.3 x 10$^{-8}$ cm/s (1.00) | 1.1 x 10$^{-7}$ cm/s (1.74) | 3.8 x 10$^{-7}$ cm/s (6.10) | 5.5 x 10$^{-8}$ cm/s (0.87) | 7.5 x 10$^{-8}$ cm/s (1.19) |
| ST$_{ot}$ | 6.3 x 10$^{-8}$ cm/s (1.00) | 1.0 x 10$^{-7}$ cm/s (1.61) | 3.5 x 10$^{-7}$ cm/s (5.60) | 6.4 x 10$^{-8}$ cm/s (1.01) | 8.2 x 10$^{-8}$ cm/s (1.31) |
| SN$_{ot}$ | 5.9 x 10$^{-8}$ cm/s (1.00) | 8.4 x 10$^{-8}$ cm/s (1.43) | 1.4 x 10$^{-7}$ cm/s (2.34) | 7.5 x 10$^{-8}$ cm/s (1.27) | 8.1 x 10$^{-8}$ cm/s (1.37) |

### Table 7 - Soil parameters used to estimate $k_M$.

| Soil sample | $e$ | $a$ | $b$ | $C_{ec}$ |
|-------------|-----|-----|-----|---------|
| SL$_{ot}$  | 0.608 | 0.00083 | -4.48 | 0.2 |
| ST$_{ot}$  | 0.735 | 295.12092 | -7.56 | 0.2 |
| SN$_{ot}$  | 0.665 | 46.77351 | -7.26 | 0.2 |

### Table 8 - Solution parameters, mesoplastic point (MP) and the resultant $k_M$ values.

| Solutions | Water | CaCl$_2$ | C$_2$H$_5$OH | HNO$_3$ | NaOH |
|-----------|-------|----------|-------------|---------|------|
| $\gamma/\mu$ (cm$^{-1}$s$^{-1}$) | 99300 | 99300 | 402165 | 99300 | 99300 |
| Sample | MP(%) | | | | |
| SL$_{ot}$ | 49.0 | 45.1 | 43.9 | 50.0 | 46.8 |
| ST$_{ot}$ | 55.8 | 53.4 | 52.7 | 56.4 | 54.9 |
| SN$_{ot}$ | 51.4 | 49.9 | 49.5 | 50.9 | 50.7 |
| Sample | $k_M$ (cms$^{-1}$) | | | | |
| SL$_{ot}$ | 6.21E-08 | 9.01E-08 | 4.12E-07 | 5.65E-08 | 7.60E-08 |
| ST$_{ot}$ | 8.34E-08 | 1.17E-07 | 5.24E-07 | 7.76E-08 | 9.51E-08 |
| SN$_{ot}$ | 6.19E-08 | 7.76E-08 | 3.32E-07 | 6.67E-08 | 6.90E-08 |

**Figure 9** - (a) Laboratory $k$ vs. theoretical $k_M$ for the “optimum bentonite content” samples; (b) Laboratory $k$ vs. $k_M$ as a function of the chemical solutions.
5. Conclusions

Four research objectives were stated in the Introduction section, and the next conclusions will be presented in the same order, as follows:

(1) “A comparison between laboratory direct and indirect methods for compatibility evaluation”. The incompatibility order was the same for both direct and indirect methods, considering all solutions and soil samples: C₂H₅OH > CaCl₂ > NaOH > HNO₃. This fact corroborates the perspective of using indirect methods as a preliminary tool for comparing different candidate soils for barrier purposes from a qualitative point of view. Small laboratory effort is needed for this last approach.

(2) “An assessment of the influence of the bentonite content on the compatibility of tropical soil samples”. It was demonstrated by Figs. 5 and 6 that the bentonite content had major influence on the decrease of the compatibility of the tropical soils for all solutions. However, exception made for the ethanol PA reagent, the compatibility indices R and kₐ close to 1 demonstrate that small proportions of bentonite, up to 12% (dry weight basis) as in this research, do not affect the soil so much.

(3) “An investigation of the influence of the laterization degree on the compatibility”. Figure 8 demonstrates that the weathering stage (laterization degree) of the tropical soil samples had minor influence on the compatibility results.

(4) “An evaluation of theoretical equations to predict the compatibility of these samples through back analysis”. In general terms, good predictions were obtained both for direct or indirect methods relative to the laboratory tests results. For indirect compatibility, these comparisons were presented in Table 4 for the bentonite sample and in Fig. 7 for the natural soil samples and mixtures considering the ethanol and calcium chloride solutions. In turn, direct compatibility comparisons were presented on Fig. 9 for all soil samples and solutions.

The issues to be addressed in future research must involve the use of other tropical soil samples comprising the many pedological taxonomies of Table 1, as well as the use of real liquids from actual disposal sites. Additionally, as the results of the present research were obtained by means of an “instantaneous” contact of the soil with the contaminant solutions, a long term analysis is suggested to evaluate possible changes on the composition and/or mechanical behavior of the compacted tropical soils.

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