Applied Clay Science 49 (2010) 187–193

Contents lists available at ScienceDirect

Applied Clay Science

journal homepage: www.elsevier.com/locate/clay

# Long term hydraulic conductivity of compacted soils permeated with landfill leachate

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ARTICLE INFO

*Article history:*
Received 3 November 2009
Received in revised form 30 April 2010
Accepted 4 May 2010
Available online 12 May 2010

*Keywords:*
Clay minerals
Bentonite
Hydraulic conductivity
Leachate
Bacteria
Pore clogging

ABSTRACT

This study evaluates the relative influence of biological, physical and chemical interactions on the percolation of leachate through compacted soil liners. The long term hydraulic conductivity of compacted silt–bentonite mixtures was measured with distilled water, landfill leachate and a nutrient solution. The soil hydraulic conductivity decreased significantly with time when the permeating liquid contained microorganisms. The decrease of hydraulic conductivity was caused by reduction of the effective porosity due to pore clogging. Pore clogging was analyzed considering physical, chemical and biological processes. The effect of microbes on the hydraulic conductivity of soil liners permeated with leachate prevailed over that produced by physical or chemical interactions. The presence of microbial activity was confirmed by direct observations of the microbial population in the permeating liquid, by microbial exopolysaccharides (EPS) encountered in the soil pores, and from inverse modeling analysis of pore bioclogging. The existence of microbes in the compacting and permeating liquids reduced up to two orders of magnitude the long term hydraulic conductivity. The results were in good agreement with the expected behavior when biofilms develop around soil particles.
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## 1. Introduction

The disposal of municipal solid waste (MSW) has the potential to impact the environment negatively. The main concern is to prevent the contamination of soil and water by the leachate that originates in the decomposition of the solid waste inside landfills (Kjeldsen et al., 2002). The volume and chemical composition of leachate depends on the water that infiltrates in the landfill, and on the chemical reactions between the solid and liquid phases, including dissolution, precipitation, ion exchange and biochemical processes.

Leachate migration from inside the landfill cell to the vadose zone is prevented by low permeability liners (Petrov and Rowe, 1997; Guyonnet et al., 2005; Touze-Foltz et al., 2006), which usually have multiple layers of compacted clay, granular filters and geosynthetics. Compacted clays or mixtures of local soils with clay are frequently used to achieve very low hydraulic conductivity barriers and prevent subsurface contamination. The hydraulic conductivity can be further reduced by the addition of bentonite to local soils to attain the values specified by international regulations ($k<10^{-7}$ cm/s) (Kayabali, 1997; Goldman et al., 1998).

The ability of compacted soil liners to restrict the movement of water and contaminants depends on particle size, void ratio, specific surface, degree of saturation, and fluid properties (Vuković and Soro, 1992; Foged and Baumann, 1999). Soil fabric, compaction energy and thixotropy are also relevant properties (Daniel and Benson, 1990; Benson and Trast, 1995). Different particle associations created during compaction generate either flocculated or dispersed soil fabrics, and are of fundamental importance in the soil hydraulic conductivity (Mitchell et al., 1965).

In the past two decades, several studies were conducted to evaluate how soil and liquid properties control the hydraulic conductivity of soil liners (Mitchell et al., 1965; Mitchell and Jaber, 1990; Gleason et al., 1997; Schmitz, 2006). In general, the hydraulic conductivity of soils decreases with increasing fine particle content (Sivapullaiah et al., 2000). At high mechanical stress levels and in the case of highly compacted soils, electrical forces have negligible effect on soil behavior and soil fabric is slightly affected by the chemical properties of the permeating liquid (Mitchell and Soga, 2005). However, hydraulic behavior of fine soils with high porosity and freshly compacted soils is highly influenced by the interaction between the pore fluid and mineral particles.

Another important property is the retention capacity of the soil, which depends on adsorption mechanisms that delay the passage of contaminants through soil liners. The adsorption of the ions present in the permeating liquid by the mineral surface is controlled by surface charge density of the particles, pH, ion concentration, ion valence, dielectric permittivity and temperature of the pore fluid (Clement et al., 1998; Aringhieri and Giachetti, 2001). The counterions in the double layer can be replaced by other hydrated cations while electroneutrality is preserved, increasing the residence time of contaminant species within soil liners (Fetter, 1993). Adsorption and ion removal mechanisms can be significantly affected when the

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doi:10.1016/j.clay.2010.05.003

permeating liquid contains several ions as in the case of polyelectrolytes and landfill leachate (Kietlińska and Renman, 2005).

The purpose of this study is to evaluate the simultaneous effect of clay content, soil porosity, chemical precipitation and microbiological properties of the permeating liquid on the long term hydraulic conductivity of compacted silt and silt–bentonite mixtures.

## 2. Materials and methods

### *2.1. Soil samples*

The soils were silt and bentonite. Table 1 summarizes relevant physical properties of these materials as well as Scanning Electron Microscopy (SEM) images and Energy Dispersive X-ray (EDX) analyses.

The silt was a typical soil from the Chaco–Pampean plain, formed by very fine sand, silt and clay particles lifted by wind and transported by eolian action (Iriondo, 1997; Zarate, 2003). These quaternary sediments cover more than 600,000 km$^2$ of the center and north-east of Argentina and are frequently used as construction materials for liners. However, the mean hydraulic conductivity of this soil after compaction is in the 10$^{-6}$ cm/s range and insufficient to be used in landfill liners (Nieva and Francisca, 2009). For this reason, bentonite or other clay minerals capable of lowering the hydraulic conductivity are commonly added to these silts to achieve the values required for the construction of containment barriers.

The bentonite contained more than 92% of sodium montmorillonite (data provided by Minarmco SA). This type of soil is frequently used in landfill liners, slurry walls and many other geoenvironmental applications due to its expansive characteristic and low hydraulic conductivity (Gleason et al., 1997).

### *2.2. Compaction and permeating liquids*

The compaction and permeating liquids were: a) distilled water (DW), b) nutrient solution (NS), and c) landfill leachate (LL). The nutrient solution contained 2% glucose, 0.1% NaCl, 0.1% yeast, 0.05% $MgSO_4$, 0.08% $K_2HPO_4$, 0.02% $KH_2PO_4$ and $7.5\times10^{-4}$% $FeCl_3$ (Dennis and Turner, 1998). The main properties of the landfill leachate are summarized in Table 2. Before testing, the leachate was filtered with a qualitative P5 filter paper to remove solid particles with diameters >5–10 µm. Yeast and naturally growing bacteria in the nutrients solution and indigenous bacteria in the leachate controlled the biological activity in these two permeating liquids.

### *2.3. Sample preparation and hydraulic conductivity tests*

Soil specimens were dried at 105 °C during 24 h. The silt fraction was then mixed with different amounts of bentonite (Table 3). The optimum moisture content for the different soil mixtures ranged from 17.6% to 19.6% and the compaction liquid content was 20% in all cases. In this case, a dispersed microstructure is expected for all specimens since they were compacted in the right side of the compaction curve (Daniel and Benson, 1990). Finally, the wet soils were compacted in rigid-wall compaction–mold permeameters using the standard proctor energy (ASTM D698 — ASTM, 2007).

Hydraulic conductivity tests were performed by the falling head technique following the ASTM D5856 standard procedure (ASTM, 2007), with hydraulic gradients between 6.5 and 2.1. The permeating liquid of each tested sample is indicated in Table 3. The hydraulic conductivity was measured every week during 15 months by monitoring the volume of liquid permeated through the specimens.

**Table 1**
Relevant properties of tested soils.

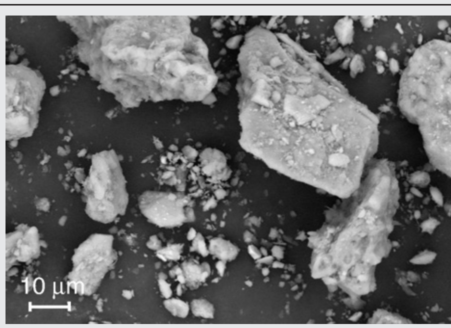
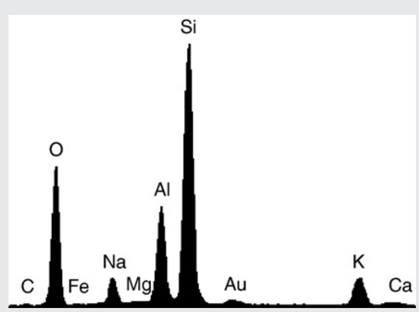
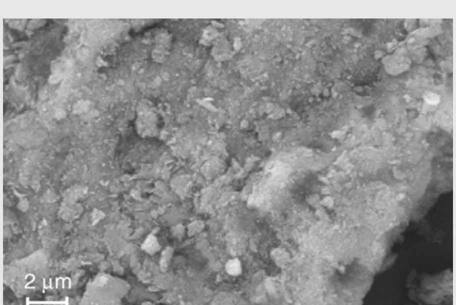
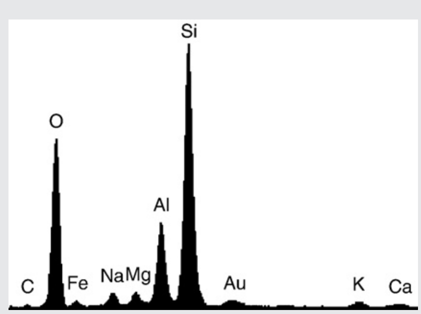
| Soil | SEM images | EDX microanalysis | Properties |
|---|---|---|---|
| Silt |   |   | Liquid limit = 27%<br>Plastic index = 2.8%<br>Particles < 74 µm = 96%<br>Particles < 2 µm = 4%<br>Specific gravity = 2.67<br>Specific surface = 2.5 m$^2$/g |
| Bentonite |   |   | Liquid limit = 285%<br>Plastic index = 240%<br>Particles < 74 µm = 100%<br>Particles < 2 µm = 80%<br>Specific gravity = 2.71<br>Specific surface = 731 m$^2$/g |

**Table 2**
Minimum and maximum values of chemical parameters of the leachate used as permeating liquid.

| Parameter | Range | Parameter | Range | Parameter | Range |
|---|---|---|---|---|---|
| BOD (mg/l) | 2500–68,500 | Total dissolved solids (mg/l) | 13,120–51,100 | Magnesium (mg/l) | 59–3860 |
| COD (mg/l) | 17,400–93,700 | Alkalinity (mg $CaCO_3$/l) | 3730–23,150 | Iron (mg/l) | 28–126 |
| TKN (mg/l) | 784–2576 | pH | 5.2–8.1 | Manganese (mg/l) | 15–96 |
| Organic N (mg/l) | 24–852 | Total hardness (mg $CaCO_3$/l) | 520–23,100 | Chromium (mg/l) | 0.14–2.20 |
| Ammonia-N (mg/l) | 61–2250 | Chloride (mg/l) | 582–4700 | Lead (mg/l) | 0.11–12.00 |
| Nitrate (mg/l) | 80–550 | Sulfate (mg/l) | 80–2000 | Cadmium (mg/l) | 0.11–0.62 |
| Total P (mg/l) | 5.20–23.50 | Sodium (mg/l) | 645–29,200 | Zinc (mg/l) | 1.0–13.5 |
| Phenols (mg/l) | 3.45–26 | Potassium (mg/l) | 70–2786 | Nickel (mg/l) | 0.03–2.1 |
| Conductivity (μS) | 21,136–68,742 | Calcium (mg/l) | 42–2880 | Copper (mg/l) | 0.20–6.60 |

### 2.4. Modeling and data analysis

The hydraulic conductivities measured in all samples were analyzed by considering the main fluid and soil properties featured by the Kozeny–Carman equation (Mitchell and Soga, 2005):

$$k = \frac{\rho g}{\mu} \frac{1}{K_n T^2 S_0^2} \left( \frac{e^3}{1+e} \right) S^3 \tag{1}$$

where $k$ is the hydraulic conductivity, $g$ the acceleration of gravity, $\rho$ the fluid mass density, $\mu$ the viscosity, $T$ the tortuosity, $K_n$ the pore shape factor, $S_0$ the wetted surface area per unit volume of particles, $e$ the void ratio and $S$ the degree of saturation.

The evolution of the hydraulic conductivity with time of permeation was analyzed by considering two possible mechanisms: a) carbonate precipitation (VanGulck et al., 2003) and b) bioclogging mechanisms (MacLeod et al., 1988; Vandevivere, 1995). The amount of carbonates in the soil specimen was determined by the $CO_2$ production with hydrochloric acid (ASTM D4373, 2007). The biological activity was confirmed by microbiological tests of the permeating liquids collected in the inlet and outlet ports. These measurements were performed by the Heterotrophic Plate Count (HPC) technique, following the American Standard Methods for the Examination of Water and Wastewater, using the pour plate method with Plate Count Agar and inoculating at 35 °C during 24 h (APHA, 1995). Bacteria detected in the NS and LL at 5 and 15 months of permeation were in the range of $10^5$ CFU/mL and $10^3$ CFU/mL, respectively.

**Table 3**
Tested samples.

| Soil fractions (%) | | Compaction fluid | Permeating liquid | Dry density (g/cm$^3$) | $\frac{e_0^3}{1+e_0}$ | kr |
|---|---|---|---|---|---|---|
| Silt | Bentonite | | | | | |
| 100 | 0 | DW | DW | 1.61 | 0.161 | 0.81 |
| 95 | 5 | DW | DW | 1.57 | 0.192 | 1.18 |
| 90 | 10 | DW | DW | 1.55 | 0.206 | 1.71 |
| 100 | 0 | NS | NS | 1.56 | 0.200 | 0.02 |
| 95 | 5 | NS | NS | 1.52 | 0.233 | 0.06 |
| 90 | 10 | NS | NS | 1.50 | 0.254 | 0.09 |
| 100 | 0 | DW | LL | 1.60 | 0.173 | 0.06 |
| 95 | 5 | DW | LL | 1.53 | 0.223 | 0.02 |
| 90 | 10 | DW | LL | 1.53 | 0.226 | 0.01 |
| 100 | 0 | NS | LL | 1.53 | 0.224 | 0.02 |
| 95 | 5 | NS | LL | 1.54 | 0.218 | 0.02 |
| 90 | 10 | NS | LL | 1.49 | 0.260 | 0.03 |
| 100 | 0 | DW | LL | 1.57 | 0.191 | 0.03 |
| 95 | 5 | DW | LL | 1.55 | 0.206 | 0.10 |
| 90 | 10 | DW | LL | 1.51 | 0.241 | 0.04 |

Note: DW = distilled water, NS = nutrient solution, LL = landfill leachate, $e_0$ = initial void ratio, $kr$ = hydraulic conductivity ratio.

Hydraulic conductivity changes related to either carbonate precipitation or bioclogging were analyzed by considering the influence of expected reductions of void ratio according to the theoretical Kozeny–Carman model (Eq. (1)), as follows:

$$kr = \frac{k_f}{k_0} = \frac{1+e_0}{1+e_f} \left( \frac{e_f}{e_0} \right)^3 \tag{2}$$

where $kr$ is the hydraulic conductivity ratio and the subscripts '0' and '$f$' represent initial and final states, respectively. Note that $e_f$ includes the effect of chemical precipitation and/or bioclogging and that Eq. (2) assumes negligible changes in liquid properties, tortuosity and specific surface during the tests. However, particles completely surrounded by biofilms may be considered as equivalent larger particles with lower specific surface (Santamarina et al., 2001). The expected change of specific surface area is generally smaller than one order of magnitude when 95% of pores are filled with biomass and can be computed as (Clement et al., 1996):

$$Mr = \frac{M_f}{M_0} = \left( \frac{e_f}{e_0} \right)^{2/3} \left( \frac{1+e_0}{1+e_f} \right)^{2/3} \tag{3}$$

where $Mr$ is the specific surface area ratio, and $M_f$ and $M_0$ the final and initial specific surface area respectively. In addition, this effect can be of relevance in the case of coarse sediments with large pores sizes but has little influence in the case of fine soils with very small pore sizes.

## 3. Results

The hydraulic conductivity of compacted silt–bentonite mixtures showed negligible time dependence when the testing liquid was DW and decreased significantly with the bentonite content (Fig. 1a). More than 5% of bentonite was needed to achieve $k<10^{-7}$ cm/s, as required by current regulations. Conversely, the samples compacted and permeated with nutrient solutions exhibited notable reductions of $k$ with permeation time (Fig. 1b).

Similar trends were observed when the specimens were compacted with DW and NS, and permeated with LL (Fig. 2). Specimens compacted with DW exhibited a time dependent behavior when permeated with leachate (Fig. 2a) in contrast to the observed trend when permeated with DW (Fig. 1a). In addition, $k$ decreased almost one order of magnitude after 12 months of testing, and the observed decrease was faster when the specimens were compacted with NS than when compacted with DW.

All samples compacted with DW showed higher hydraulic conductivity than those compacted with NS after 12 months of testing, regardless of bentonite content. However, in both cases $k$ clearly decreased with increasing bentonite content (Figs. 1a and 2a), and also showed some variability with permeation time for samples compacted with NS (Figs. 1b and 2b).

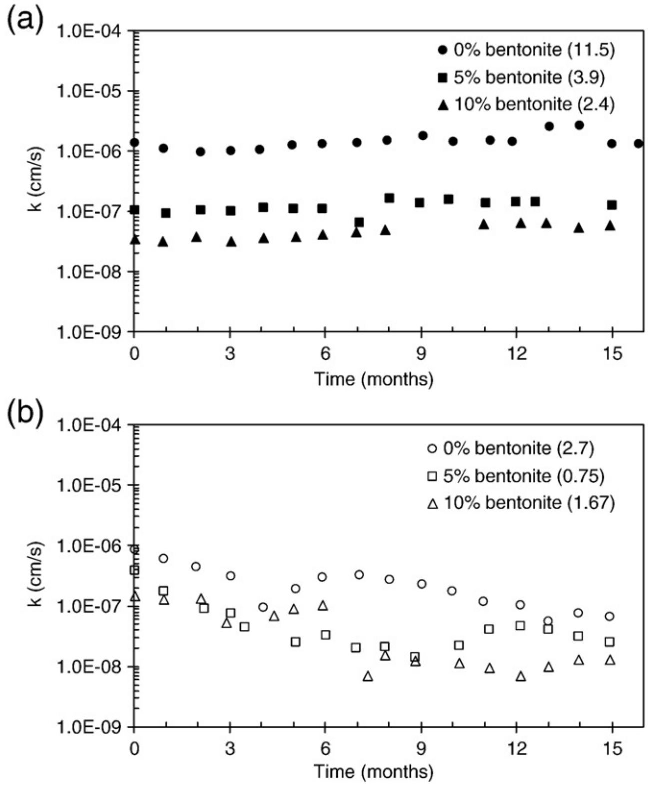



**Fig. 1.** Influence of time on the hydraulic conductivity of silt–bentonite mixtures: a) samples compacted and permeated with distilled water; b) samples compacted and permeated with nutrient solution. Numbers between brackets indicate the pore volume of flow after 15 months of permeation.

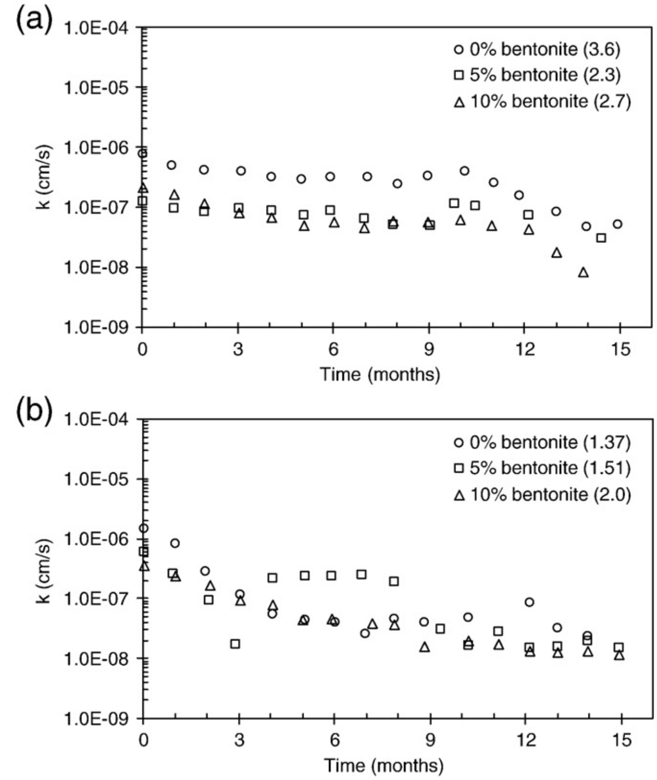



**Fig. 2.** Influence of time on the hydraulic conductivity of silt–bentonite mixtures permeated with leachate: a) samples compacted with distilled water; b) samples compacted with nutrient solution. Numbers between brackets indicate the pore volume of flow after 15 months of permeation.

The initial porosity ($n_0$) of compacted silt–bentonite mixtures had negligible influence on *kr* obtained after 15 months of testing (Fig. 3). In addition, there was a very small influence of bentonite content on *kr* when the specimens were compacted with the nutrients solution or permeated with this solution or leachate. *kr* became close to one when the permeating liquid was DW, given that $k_0 \cong k_f$ (Fig. 1a), and fell between $10^{-2}$ and $10^{-1}$ for the specimens permeated with LL. These results confirmed the importance of liquid properties on the expected final hydraulic conductivity of compacted earthen liners.

Optical microscopy images confirmed the presence of bacteria and yeast in the liquid collected in the outlet port of all the samples tested with LL and NS (Fig. 4a). In addition, Fig. 4b shows microbial exopolysaccharides (EPS) detected in SEM images of the soil after permeation confirming the presence of microbial activity within soil pores.

## 4. Discussion

### *4.1. Soil properties*

In this study all the specimens were compacted with moisture content higher than the optimum and consequently disperse fabrics were expected. In this condition, the measured hydraulic conductivity reached its lowest possible value, assuming no changes of compaction energy and method (Mitchell and Soga, 2005).

The initial void ratios ranged from 0.66 to 0.81 and could not be responsible of any significant change of the hydraulic conductivity, $k \propto e^3/(1+e)$, as theoretically shown by the Kozeny–Carman equation. In addition, the initial soil saturation ($S$) between 65% and 70% increased to >85% during the test. $k$ decreased with the permeation time even there were more pores participating in the flow due to the higher saturation.

Observed permeability changes could be related to: a) expansion/shrinkage of expansive minerals, b) mineral clogging, and c) bioactivity. The replacement of the $Na^+$ ions of the sodium bentonite by more highly charged cations and/or the increase of ionic concentration usually decrease the double-layer thickness, and rise the soil hydraulic conductivity (Schmitz, 2006). The landfill leachate, characterized by a high ionic concentration, should then increase the

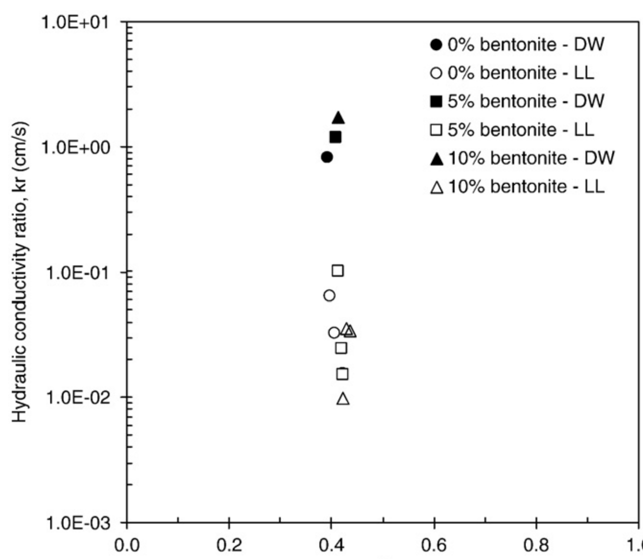



**Fig. 3.** Relationship between the hydraulic conductivity ratio after 15 months of permeation and the initial soil porosity ($n$). Permeating liquids: DW = distilled water, NS = nutrient solution, LL = landfill leachate.

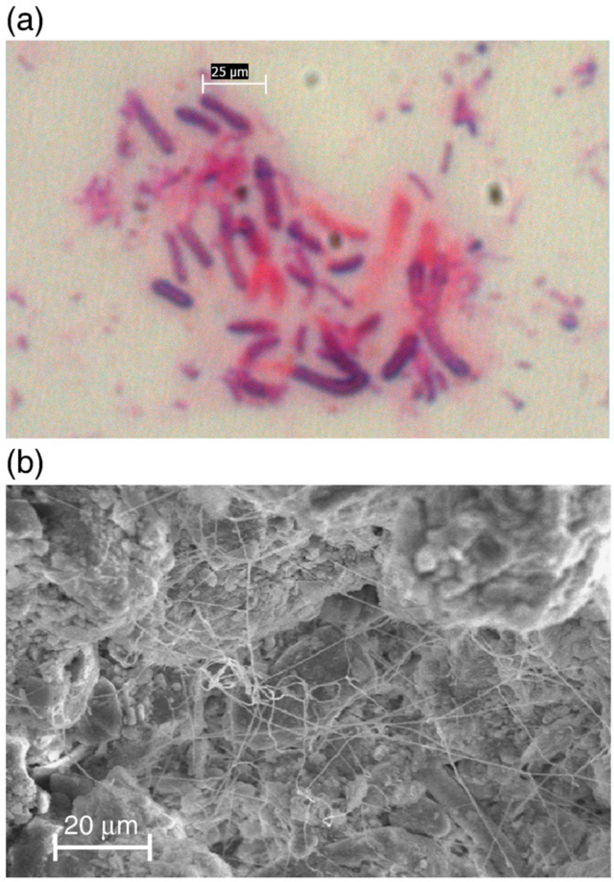



**Fig. 4.** a) Typical microorganisms identified in the permeating liquid collected in the outlet port, b) SEM image of the specimen permeated with leachate. Filament-like structures are microbial exopolysaccharides secreted by microbes inside the soil pores.

soil hydraulic conductivity when flowing though soil pores. However, this mechanism had negligible effect on the experimental tests since it should be relevant only in high porosity or freshly compacted soils (Mitchell and Soga, 2005). Conversely, $k$ decreased with the permeation time indicating that other mechanisms (i.e. pore clogging) were controlling the liquid displacement through the soil pores.

### 4.2. Pore clogging and bioactivity

Fluid displacement inside soil pores could also be restricted by pore clogging mechanisms. Physical clogging had a negligible effect because the hydraulic conductivities reported in Figs. 1b and 2 decreased with time even though the filtered leachate and nutrient solution used as permeating liquids did not contain solid particles that could block the soil pores. Chemical clogging was produced by precipitation partially occupying void spaces within the sample (e.g. carbonate precipitation). This mechanism was previously reported by VanGulck and Rowe (2004) when different soils were permeated with leachate. The natural carbonate content of the samples was 30 mg/g (by mass of dry soil). After permeation, measured values fell between 33 and 41 mg/g, and the concentration of precipitated carbonate was close to 11 mg/g. Since the tested specimens had a total volume $V = 950\ cm^3$ and the mean soil porosity was $n = Vv/V = 0.42$, the maximum amount of precipitated carbonate could reduce the porosity by only 1.5% and could not be responsible of the observed decrease of the hydraulic conductivity.

The presence of nutrients was responsible for the formation and stimulation of yeast and bacteria colonies that partially blocked the soil pores (Rebata-Landa and Santamarina, 2006). The decrease of $k$ due to bioactivity can be related to the presence of biofilms and associated bioclogging mechanisms controlled by the relative size of microorganisms respect to pore and throat sizes.

There are several theoretical and empirical models that can be used to estimate the influence of microorganisms on the decrease of $k$ (Fig. 5). These models consider different mechanisms affecting hydraulic conductivity, which are associated with the presence of uniform biofilm accumulation (Vandevivere, 1995; Seki and Miyazaki, 2001; Thullner et al., 2002), cell aggregate clogging mechanisms (Ives and Pienvichitr, 1965; Clement et al., 1996) and the development of colonies (Thullner et al., 2002). The contribution of bacteria in reducing the hydraulic conductivity depends on the final void ratio ($e_f$) which is computed from $kr$, the initial void ratio ($e_0$) and Eq. (2). Furthermore, the volume of bacteria filling the pores ($e_b$) and bacteria biovolume ($B$) become:

$$e_b = e_0 - e_f \tag{4}$$

$$B = \frac{e_b}{e_0} \tag{5}$$

The Vandevivere (1995), Seki and Miyazaki (2001) and Thullner et al. (2002) models, implemented by means of least-squares fitting, and the Clement et al. (1996) model, which contains no empirical or fitting parameters, are shown in Fig. 5. The obtained trends showed good agreement between the experimental results and the values predicted with the biofilm models. Lower $k_f$, represented by flow vectors, were obtained at higher $B$ since the biofilms restricted fluid displacement.

A multiple regression analysis was performed with the purpose of verifying the relevance of bioactivity and relevant physical properties of soils on the hydraulic conductivity ratio $kr$. Fig. 6 presents the computed and measured final hydraulic conductivities for the silt-bentonite mixtures permeated with the LL and NS. The model

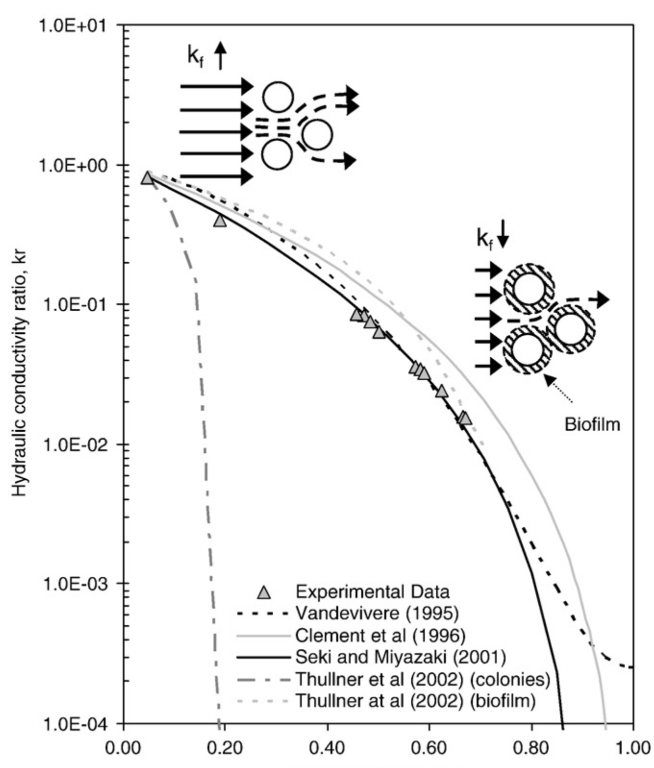



**Fig. 5.** Influence of the volume fraction of pore space occupied by bacterial colonies on the hydraulic conductivity ratio.

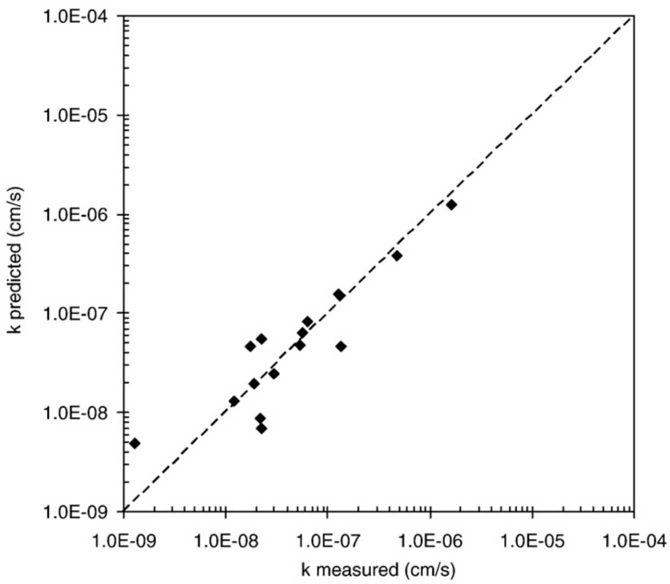



**Fig. 6.** Measured and predicted long term hydraulic conductivities of silt–bentonite mixtures compacted with distilled water and permeated with leachate. Numerical simulations are performed by means of multiple regression analysis ($R^2 = 0.814$).

parameters, bentonite content, soil dry density and void ratio function are presented in Table 3. The bioactivity was considered by the ratio between the size of the microbial cells in the permeating liquid and the bacteria size. The size ratios were 0, 1 and 10 for distilled water, leachate and nutrient solution given that distilled water had not bacteria and yeast inoculated in the nutrient solution was approximately ten times larger than bacteria. The agreement between the measured and computed reduced hydraulic conductivity confirmed that the presence of microorganisms controlled the liquid displacement in soils permeated with leachate.

## 5. Conclusions

The hydraulic conductivity of compacted silts decreased with the bentonite content when permeated with distilled water. In this case, the permeation time had negligible influence on *k*, disregarding the bentonite content.

Physicochemical interactions including changes in the double-layer thickness and chemical precipitation of carbonates had negligible effect on the hydraulic conductivity of highly compacted silt–bentonite mixtures permeated with leachate.

Inoculating bacteria and yeast in the compacting and permeating liquid reduced significantly the hydraulic conductivity of compacted silt–bentonite mixtures. This decreased *k* with the permeation time, by a factor of $10^2$, between the specimens compacted using distilled water and compacted using a nutrient solution. The presence of biomass in the compacting liquid more strongly decreased the long term *k* than did the presence of bentonite.

The effective porosity, which contributed to fluid flow through soil liners, was significantly reduced due to microorganisms growth inside the soil pores. Several theoretical solutions were used to compute the hydraulic conductivity ratio caused by bioclogging. The experimental data confirmed the biofilm formation. Thus, a potential way to achieve low *k* values for liners in situ could be to compact them using a nutrient solution.

## Acknowledgements

This research was partially supported by the National Agency for the Promotion of Science and Technology (ANPCyT, research project number PICT 2006-822) and the SECyT-UNC (research project number 05/M118). D.A.G. thanks the support given by SECyT-UNC during this research. The authors thank P.M. Nieva and M.A. Montoro for their help with the measurements and the anonymous reviewers for their useful comments.

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