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The role of chemico-osmosis in the performance assessment of bentonite-based contaminant barriers

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Abstract. Although conservative estimates of the containment performance of bentonite-based barriers can be obtained from the classical advective-diffusive transport theory, the semipermeable properties of the bentonite component can determine a significant reduction in the migration rate of contaminants at low salt concentrations (< 100 mM). After introducing the transport equations for a semipermeable porous medium, the paper presents the laboratory testing apparatuses and procedures that allow the chemico-osmotic efficiency coefficient, ω, and the osmotic effective diffusion coefficient, D_{ω}^{*}, of bentonites to be experimentally assessed. Finally, the advantages that derive from the ability to model the semipermeable behaviour of bentonites in the design of geoenvironmental barriers are discussed with the aid of a calculation example, which considers the use of geosynthetic clay liners for the bottom lining of waste disposal facilities.

1. Introduction

Water can flow through clay soils that are characterized by a high specific surface and an electric net charge (e.g. bentonites) in response not only to a gradient in the total hydraulic head, but also to gradients in the chemical composition (chemico-osmosis). As a result, when the volumetric flow is hindered, a pressure difference is generated at the boundaries of such clay soils. The coupled phenomenon is observed when salt concentration changes are produced by a water pressure-driven flow [1-4]. A porous medium with such properties is termed “permiselective” or “semipermeable”, as these phenomena are generated by its ability to restrict accessibility of solutes to its pores. This restriction occurs in bentonites when the pore sizes between individual clay particles are sufficiently small, so that electrostatic repulsion of the ions results from the interaction with the negatively charged clay particles [5-12].

The semipermeable behaviour is expected to have a significant influence on the performance of water and/or contaminant barriers constituted wholly or partially by bentonite, such as geosynthetic clay liners (GCLs), compacted clays, clay or sand-bentonite mixtures, and soil-bentonite cutoff wall backfills [13-16]. For this reason, understanding the physical mechanisms that govern the transport properties of bentonites is fundamental to assess the ability of such materials to limit the migration of water and contaminants in a number of geoenvironmental applications, including landfills, waste impoundments and groundwater remediation systems. This paper aims to illustrate the practical relevance of chemico-osmosis in different applications involving bentonites by presenting the

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phenomenological and physical approaches that have been developed for modelling the coupled transport of water and solutes through semipermeable porous media.

2. Transport equations for semipermeable porous media

The phenomenological framework to describe coupled fluxes of water and ions through semipermeable porous media, such as bentonites, is based on the principles of thermodynamics of irreversible processes for non-equilibrium systems [17-20]. For a bentonite layer in contact with a single salt (single cation, single anion) solution, the volumetric liquid flux, q, and mass salt flux, J_s, may be written as follows:

$$q = -\frac{k}{\gamma_w}\left(\frac{\mathrm{d}u}{\mathrm{d}x} - \omega\frac{\mathrm{d}\Pi}{\mathrm{d}x}\right) \tag{1a}$$

$$J_s = (1-\omega)qc_s - nD_\omega^*\frac{\mathrm{d}c_s}{\mathrm{d}x} \tag{1b}$$

where k is the hydraulic conductivity, γ_w is the water unit weight (9.81 kN/m^3), u is the hydraulic pressure, ω is the reflection coefficient, which is also known as chemico-osmotic efficiency coefficient, $\Pi = \nu RTc_s$ is the osmotic pressure, ν is the sum of the stoichiometric coefficients of cation and anion species, R is the universal gas constant (8.314 J·mol^{-1}·K^{-1}), T is the absolute temperature, c_s is the molar concentration of the salt, n is the bentonite porosity and D_ω^* is the osmotic effective diffusion coefficient.

Even though the thermodynamics of irreversible processes provides a consistent formulation of the transport equations, the number of laboratory tests that are necessary to characterise a single bentonite clay is high, as the phenomenological coefficients k, ω and D_ω^* are unknown functions of the state variables c_s and n. The latter drawback of the phenomenological approach can be obviated by a proper identification of the electrochemical interactions that establish between the solid phase and the ions dissolved in the pore solution, as proposed by Dominijanni and Manassero [21] on the basis of a model for bentonite structure at microscale, which is characterized by a slit-like geometry of pores. This physical interpretation of the transport parameters of bentonites was derived from an upscaling of the Navier-Stokes equation for the volumetric liquid flux and the Nernst-Planck equations for the ion mass fluxes, and using the Donnan equations to relate the hydraulic pressure and the ionic concentrations of the pore solution to the hydraulic pressure and the ionic concentrations of the external bulk solutions in contact with the porous medium at its boundaries. In such a way, the reflection coefficient can be related to the bentonite void ratio, e, the salt concentration, c_s, and the solid charge coefficient, $\bar{c}'_{sk,0}$, which accounts for the fraction of the surface charge density of montmorillonite (i.e. the main mineralogical component of bentonites) that electrostatically interacts with the mobile ionic species [22]. A linear relationship was proven to exist, under the assumption that pore-scale variations in pressure, ion concentration, and water velocity within the porous medium are negligible, between the osmotic effective diffusion coefficient and the reflection coefficient:

$$D_\omega^* = (1-\omega)\tau_m D_{s,0} \tag{2}$$

where τ_m is the matrix tortuosity factor and $D_{s,0}$ is the free-solution or aqueous-phase diffusion coefficient of the salt.

The original formulation of the above physical model, as proposed by Dominijanni and Manassero [21], pointed out the relation between $\bar{c}'_{sk,0}$ and k and the bentonite microstructure, consisting of non-conductive pores within the tactoids (i.e. stacks of parallelly aligned montmorillonite unit layers) and conductive pores between the tactoids, where a single state parameter, referred to as the average number of lamellae per tactoid, $N_{l,AV}$, allows the soil fabric to be completely defined (see figure 1).

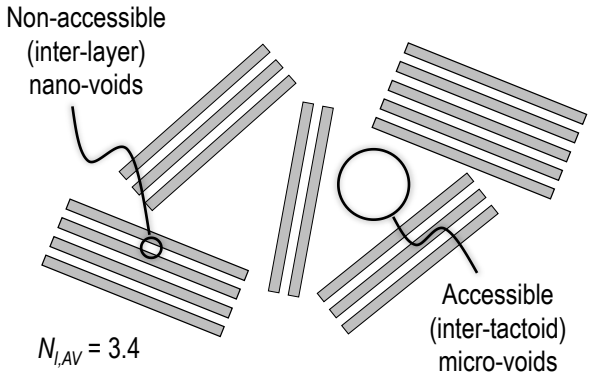

Figure 1. Schematic view of the bentonite microstructure according to the dual-porosity model proposed by Dominijanni and Manassero [21].

Flocculation phenomena, characterized by an increase in $N_{l,AV}$, were shown to be able to induce a decrease in $\bar{c}'_{sk,0}$ and a large increase in k. In a series of successive works, the physical model was further improved through the introduction of the so-called fabric boundary surface (FBS), whereby the average number of lamellae per tactoid, $N_{l,AV}$, is related to the salt concentration, c_s, and the micro-void ratio, e_m, which in turn is obtained by subtracting the void space between the platelets of the tactoids from the total void space [14, 15, 23, 24]. When significant changes occur in the salt concentration or in the confining stress, the use of FBS allows the effects of bentonite fabric changes on the macroscopic transport properties to be taken into account.

3. Laboratory assessment of the bentonite semipermeable properties

The main components of the testing apparatus which was used by Malusis and Shackelford [25], Dominijanni et al. [26, 27] and Musso et al. [28], among others, to measure the reflection coefficient and the osmotic effective diffusion coefficient of bentonites are schematically illustrated in figure 2, and include the osmotic cell, the flow pump system, the differential pressure transducer and the data acquisition system. Moreover, figure 3 shows a pictorial view of the laboratory equipment.

The cell consists of a modified rigid-wall permeameter, in which the top piston and the bottom pedestal are equipped with three ports each: two enable the different solutions to circulate through the top and the bottom porous stones, with the aim of establishing a constant concentration gradient across the specimen. The third port is installed in both the top piston and the bottom pedestal to allow the differential pressure across the specimen to be measured. The flow pump system, which consists of a dual-carriage syringe pump and two stainless steel accumulators, prevents volumetric flux through the specimen by simultaneously injecting into and withdrawing from the porous stones the same volume of solution.

The chemico-osmotic test is performed according to the procedure proposed by Malusis and Shackelford [25]: a solution containing a known electrolyte concentration is circulated in the top porous stone, while a solution with lower concentration (e.g. deionised water) is circulated in the bottom porous stone. The concentration difference across the specimen is maintained constant over time by continuously infusing the two liquids at the specimen boundaries. The electrolyte concentration of the withdrawn fluxes (i.e. from the top and bottom porous stones) is monitored by sampling the solution contained in the accumulators. Therefore, on account that the syringes move at a known rate, the diffusive solute flux can be calculated from the measured changes in electrolyte concentration at the specimen boundaries.

The measured global value of the reflection coefficient, ω_g, which corresponds to the integral mean value of ω that is calculated with respect to the boundary salt concentrations, is then given by [29, 30]:

$$\omega_g = \left(\frac{\Delta u}{\Delta \Pi}\right)_{q=0} = \frac{1}{\Delta c_s}\int_{c_b}^{c_t} \omega \, dc_s \tag{3}$$

where Δu is the steady state difference in hydraulic pressure across the specimen, $\Delta c_s = c_t - c_b$ is the difference in salt concentration across the specimen, and c_t and c_b are the salt concentrations of the external bulk solutions in contact with the top and bottom specimen boundaries, respectively.

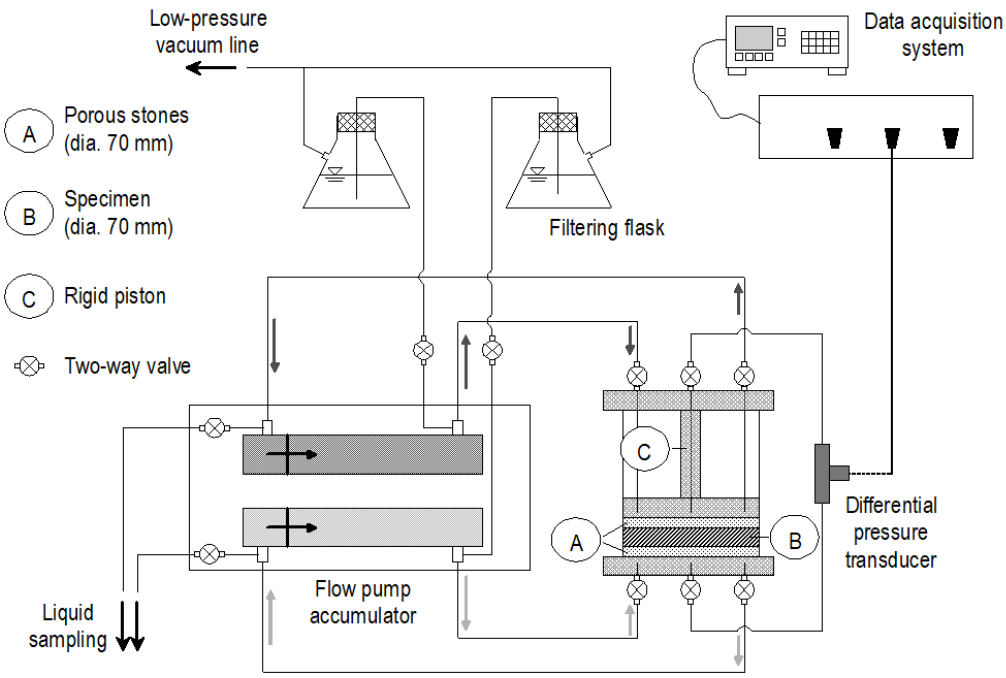

Figure 2. Schematic view of the chemico-osmotic testing apparatus (not to scale).

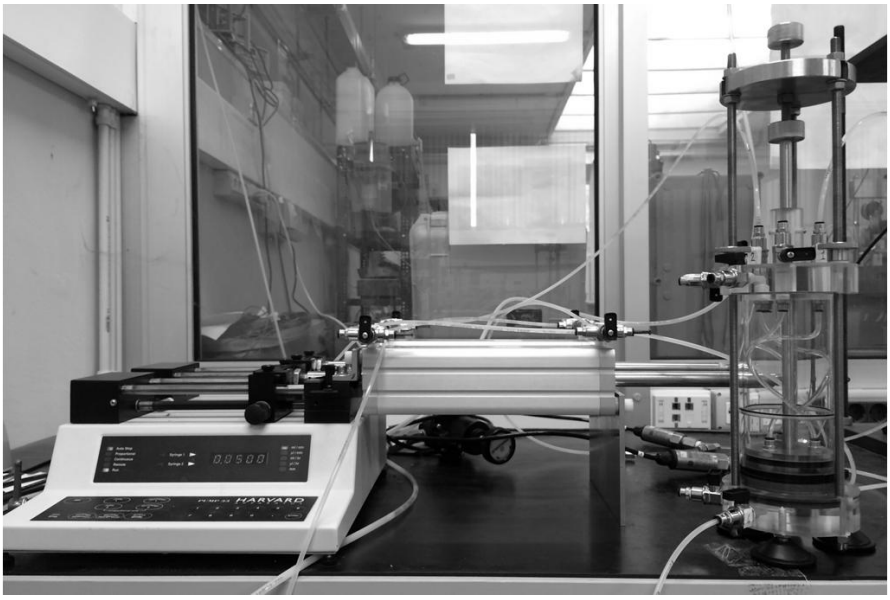

Figure 3. Pictorial view of the chemico-osmotic testing apparatus.

Once the steady state condition for the salt molar flux has been reached, the global osmotic effective diffusion coefficient, $D^*_{\omega g}$, which corresponds to the integral mean value of D^*_{ω} that is calculated with respect to the boundary salt concentrations, is determined as follows [26, 27]:

$$D^*_{\omega g} = \frac{L}{n}\left[\frac{(J_s)_{ss}}{\Delta c_s}\right]_{q=0} = \frac{1}{\Delta c_s}\int_{c_b}^{c_t} D^*_{\omega}\, dc_s \tag{4}$$

where L is the length of the specimen and $(J_s)_{ss}$ is the salt molar flux measured under steady state conditions.

Several test results have been interpreted according to the previously illustrated theoretical model by assuming, as a first approximation a constant value of $\bar{c}'_{sk,0}$. A dimensionless reference salt concentration, η_ω, calculated as follows [27]:

$$\eta_\omega = \frac{2ec_{s,\omega}}{\bar{c}'_{sk,0}}, \tag{5}$$

where $c_{s,\omega}$ is the so-called reference salt concentration that is defined as the salt concentration value, bounded between the extreme values c_t and c_b, which satisfies the following condition for any given testing stage:

$$\omega(c_{s,\omega})\cdot\Delta c_s = \int_{c_b}^{c_t} \omega\, dc_s, \tag{6}$$

has been introduced to compare the results obtained on different bentonites and using different boundary conditions. The good agreement between experimental data and model predictions that has been found is shown in figures 4 and 5.

Due to the narrow range of salt concentrations that was investigated in the considered studies, the measured values of ω_g have been interpreted through calibration of a constant value of $\bar{c}'_{sk,0}$, which varies between 30 and 48 mM for the granular bentonite GCL tested by Malusis and Shackelford [25, 33], Malusis et al. [31] and Shackelford et al. [32], suggesting the formation of an aggregated microstructure that minimises the fraction of the surface charge density interacting with the ionic species in the pore solution. On the contrary, $\bar{c}'_{sk,0}$ varies between 80 and 110 mM for the powdered natural sodium bentonite tested by Dominijanni et al. [26, 27] and Musso et al. [28], and increases up to 225 mM in the case of the homoionised sodium montmorillonite tested by Kemper and Rollins [5], preliminarily separated by its coarser fraction (> 2 μm) by sedimentation. The latter specimen is thus expected to be characterised by a dispersed fabric of montmorillonite unit layers, corresponding to extremely low values of $N_{l,AV}$.

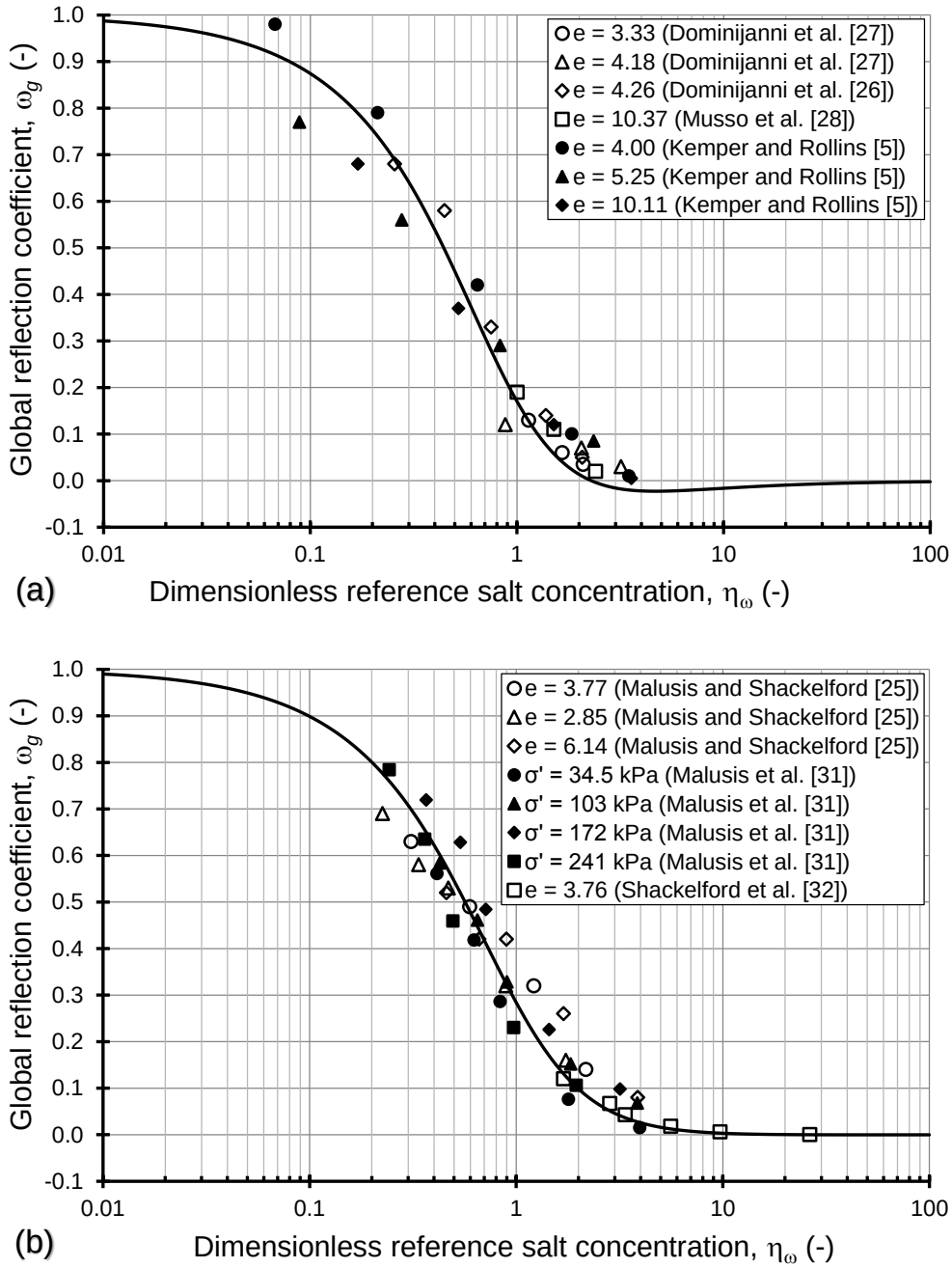

Figure 4. Measured values of the global reflection coefficient, ω_g, in equilibrium with (a) sodium chloride and (b) potassium chloride solutions, and theoretical interpretation based on the physical model proposed by Dominijanni and Manassero [21] (continuous line).

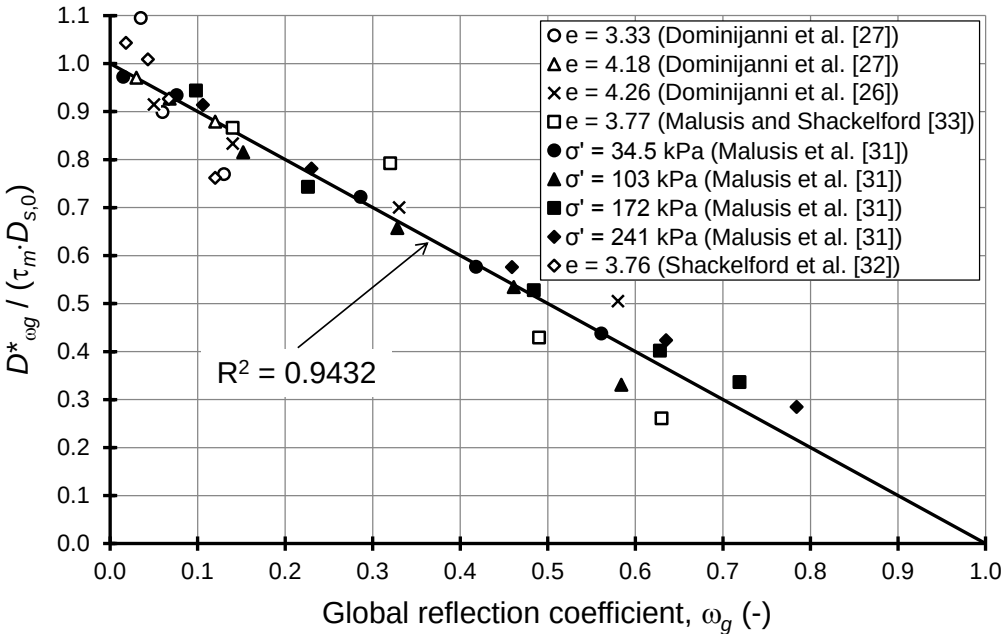

Figure 5. Measured values of the global osmotic effective diffusion coefficient, $D^*_{\omega g}$, versus the corresponding measured values of the global reflection coefficient, ω_g, and theoretical interpretation based on the physical model proposed by Dominijanni and Manassero [21] (continuous line).

4. Practical significance of the chemico-osmotic test results

The advantages that arise in the design of geoenvironmental barriers from the experimental measurement of the semipermeable properties of bentonites and the ability to model the coupled solvent and solute fluxes are highlighted herein with the aid of a calculation example. The steady state analysis of water and contaminant transport is conducted for the geosynthetic clay liner (GCL) shown in figure 6, which is used as the bottom liner of a waste disposal facility. The GCL is assumed to contain the natural sodium bentonite tested by Dominijanni et al. [26], which is characterised by a solid charge coefficient $\bar{c}'_{sk,0}$ = 90 mM, a hydraulic conductivity $k = 8 \cdot 10^{-12}$ m/s and a matrix tortuosity factor $\tau_m = 0.31$. The contaminant of interest consists of sodium chloride, the height of the ponded leachate overlying the bentonite liner is h_p = 30 cm, and L_{GCL} = 1 cm is the thickness of the hydrated GCL. A perfectly-flushing boundary condition is hypothesised within the underlying aquifer: such a calculation approach can be considered conservative, as it maximises the diffusive salt flux.

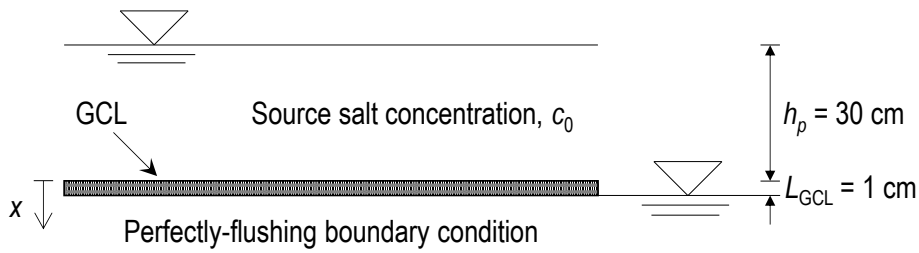

Figure 6. Reference scenario for the steady-state analysis of contaminant transport.

The calculated steady state liquid and salt fluxes are plotted in figure 7 versus the contaminant concentration in the leachate drainage layer. If the semipermeable behaviour of the GCL is taken into account, a significantly better agreement between experimental data and model predictions is observed. Furthermore, the contaminant mass flux is noticed to decrease relative to the case of absence of semipermeable behaviour, due to the simultaneous occurrence of chemico-osmosis, as evidenced by a resulting liquid flux that is directed upward (i.e. opposite to the hydraulic component of the liquid flux), and restricted diffusion [2, 16, 30, 31, 34].

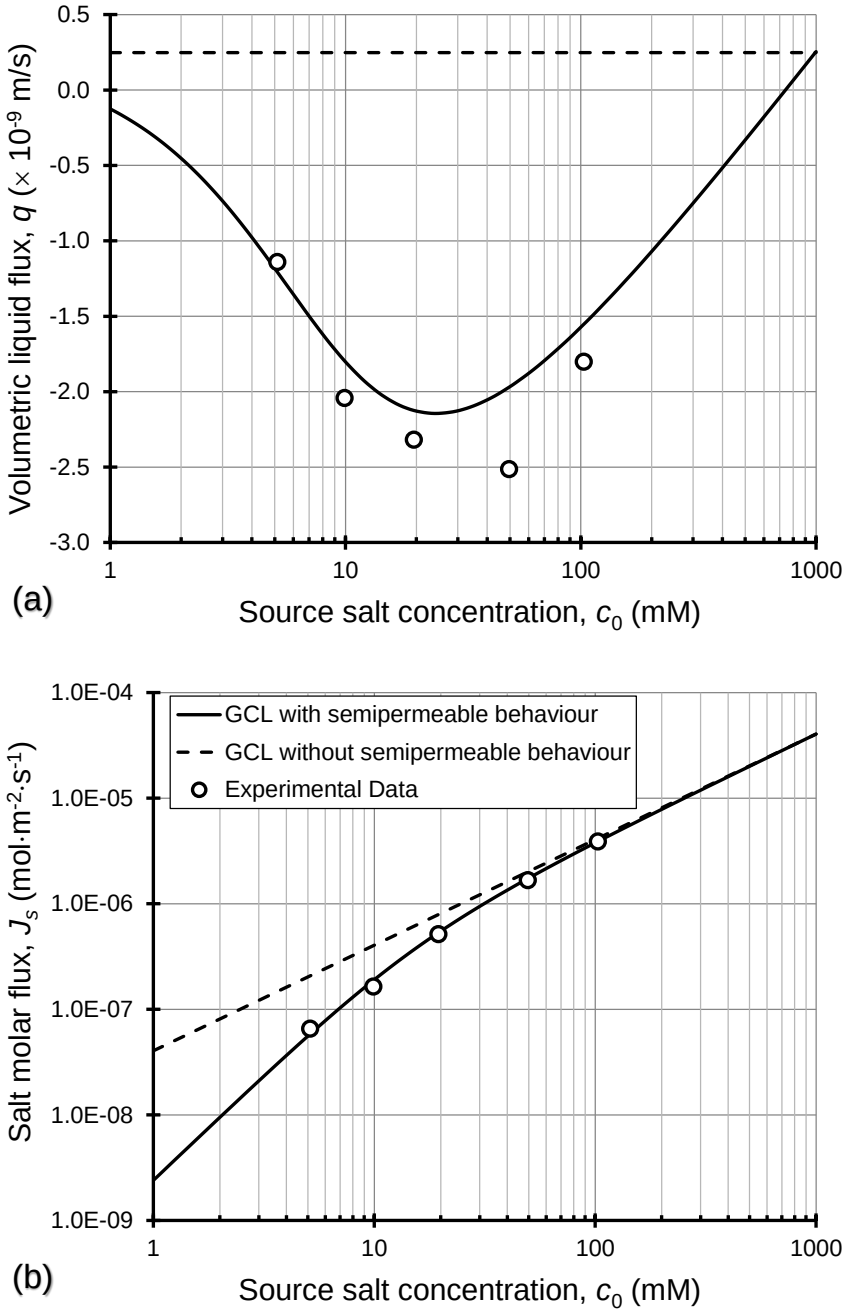

Figure 7. Theoretical simulation of (a) the leakage rate and (b) the contaminant mass flux through a GCL used as the bottom liner of a waste disposal facility.

5. Conclusions

A theoretical approach to model the coupled fluxes of solvent and solutes through semipermeable bentonite-based barriers, as well as the testing apparatuses and procedures to measure the bentonite semipermeable properties, have been presented. The ability of such a theoretical framework to properly simulate the macroscopic effect of the pore-scale electrical interactions has been verified through interpretation of laboratory test results for a variety of bentonite types (untreated vs homoionised specimens), grain size distributions (granular vs powdered specimens), void ratios and chemicals contained in the equilibrium bulk solutions. Finally, the importance of accounting for the semipermeable behaviour of bentonites in the design of contaminant barriers has been discussed with the aid of a calculation example, focusing on the use of GCLs as the bottom liners of landfills. However, the results of this study pertain to a wider spectrum of geoenvironmental applications, ranging from the slurry cutoff walls for the remediation of polluted areas to the high-density bentonite buffers for the deep geological disposal of radioactive wastes.

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