Migration Parameters of Chloride and Potassium in GCLs from Diffusion and Column Tests

Francesco Mazzieri [1*], *Davide* Bernardo [2], *Evelina* Fratalocchi[1]

[1]Università Politecnica delle Marche, SIMAU Department, 60131 Ancona, Italy
[2] SPS Mobile, 60131 Ancona, Italy

Abstract. Prediction of contaminant transport through barrier systems requires entering the solute transport parameters (e.g. diffusion coefficients and retardation factors) into analytical or numerical transport models. In low permeability hydraulic barriersfor containment applications (e.g. GCLs), molecular diffusion often is the dominant solute transport mechanism. The paper describes the determination of solute transport parameters of a GCL by means of through-diffusion (diffusive transport only) and column tests (diffusive and advective transport). Diffusion tests were performed at two stress levels (70 kPa and 241 kPa) while a column test was performed at 70 kPa, using a 20 mM KCl solution. Very close transport parameters for Cl^- and K^+ were obtained by the different testing approaches. The results confirmed the dominant effect of diffusion in transport through GCLs and, besides the reduction in steady-state osmotic efficiency shown in previous studies, also highlighted the enhancing effect of consolidation on effective diffusion coefficients and retardation factors.

1 Introduction

Geosynthetic Clay Liners, (GCLs) consisting of a thin layer of bentonite clay incorporated between two geotextiles have been widely used as hydraulic and pollutant barriers. The content of sodium montmorillonite is the key factor for the characteristically low hydraulic conductivity of GCLs. Sodium bentonite can also exhibit considerable adsorption of cations, including heavy metals, owing to its large cation exchange capacity (CEC 80-100 meq/100g). In containment scenarios, provided the typically low hydraulic conductivity of GCLs be preserved, molecular diffusion often represents the dominant solute transport mechanism [1]. Owing to the sodium bentonite, GCLs have been also shown to behave as semipermeable membranes, i.e., restrict the transport of solutes while a flow of water (chemical osmosis) is induced in the direction opposite to that of the chemical gradient (e.g., [2]). The extent to which a clay or GCL exhibits semi-permeable membrane behaviour has been typically quantified by the chemico-osmotic efficiency ω or "reflection" coefficient ([3], [4]). In addition to the potential reduction of the liquid flux owing to the onset of chemical osmosis, solute restriction also reduces the effective diffusion coefficient and, therefore, the total mass flux of the solute migrating through the GCL (e.g., [4], [5]).

Prediction of contaminant transport through barrier systems, including GCLs, requires considering the relevant mechanisms and entering the solute transport parameter (e.g., diffusion coefficients and retardation factors) into analytical or numerical transport models. One of the factors that may affect the reliability of predictions is the stress state, which influences the transport parameters of clays and GCLs. The diffusion coefficient of Na^+ and Cl^- of various GCLs has been shown to decrease approximately linearly with the bulk void ratio, e_b, for the typical range of e_b expected in the field as a function of the stress state and hydration conditions [6]. The membrane efficiency of GCLs has been shown to decrease with the increase of the consolidation effective stress (σ') applied prior to chemico-osmotic testing. For example, an increase in σ' from 34.5 kPa to 241 kPa correlated with an increase in membrane efficiency from 0.05 to 0.18 when a 20 mM KCl solution was used as source solution [7]. Therefore, a realistic assessment of solute transport parameters, including those relevant to semipermeable membrane behaviour, requires simulating the field conditions (e.g. stress state and void ratio).

The paper describes the determination of transport parameters of two solutes (Cl^- and K^+) on a needle-punched GCL by means of through-diffusion and column tests. Through-diffusion tests were carried out by means of a recently developed chemico-osmotic diffusion testing apparatus. Column tests were performed by means of flexible wall permeameters. The purpose of the study was: 1) to assess the chemico osmotic efficiency, solute diffusion coefficients and retardation factors of the GCL consolidated to different effective stress; 2) to compare the solute transport parameters of the GCL obtained under pure diffusion conditions (i.e., in through-diffusion tests) with those obtained in the presence of hydraulic flow (advection plus diffusion) in view of the use in transport models.

* Corresponding author: f.mazzieri@staff.univpm.it

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2 Materials and Methods

2.1 Materials

The GCL used in this study is needle-punched. It consists of an upper nonwoven polypropylene geotextile (300 g/m^2) and a carrier woven geotextile (200 g/m^2), encapsulating a nominal 4200 g/m^2 of powdered natural sodium bentonite (oven-dried mass). The upper geotextile is impregnated with powdered bentonite (800 g/m^2). The bentonite has a cation exchange capacity, CEC of 98 meq/100g, with sodium (77 meq/100 g) and calcium (18 meq/100 g) and magnesium (8 meq/100g) as the main adsorbed cations. The liquids utilized were distilled water (DW) and an electrolyte solution 20 mM KCl prepared by dissolving analytical grade solid KCl in DW.

2.2 Through-diffusion tests

The diffusion tests were carried out by means of the chemico-osmotic diffusion testing apparatus shown in Fig. 1. For clarity, the main features of the apparatus are briefly described herein. In accordance with the closed-system approach for chemo-osmotic diffusion testing [4] the principle is to induce a steady-state chemical gradient across a saturated soil/GCL specimen, while preventing the hydraulic flow. In case of membrane behaviour, a differential pressure will develop across the specimen, measured by means of pressure transducers.

The soil or GCL specimen is accommodated in a rigid wall plexiglass cell consisting of two chambers hydraulically separated by a sealed rigid piston. The mould hosting the specimen is 100 mm in diameter and 100 mm in height. The soil/GCL specimen is contained between two porous stones. The chemical gradient is induced across the soil by circulating two electrolyte solutions at different concentrations through the porous stones at the same volumetric rate, such that the solution is infused at one side of the porous plate and withdrawn at the other side.

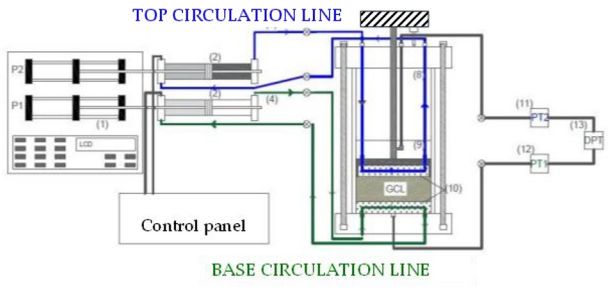


Fig. 1. Chemico-osmotic diffusion testing apparatus (modified from [8])

The tests described herein were conducted with the top piston blocked to prevent volume changes of the specimen, according to the conventional approach to chemico-osmotic diffusion testing. In case of observed membrane behaviour, the chemico-osmotic efficiency coefficient, ω is given by [4]:

$$\omega = \left(\frac{\Delta P}{\Delta \pi}\right) \quad (1)$$

where ΔP is the measured differential pressure across the specimen, $\Delta \pi$ is the theoretical chemico-osmotic pressure difference across an "ideal" semipermeable membrane, which in case of dilute solutions of a strong electrolyte can be computed using the van't Hoff equation:

$$\Delta \pi = \nu R T \Delta C \quad (2)$$

where ν is the number of ions per salt molecule, R is the universal gas constant [8.314 J mol^{-1}°K^{-1}], T is the absolute temperature, ΔC represents the concentration difference of the salt across the membrane. While it is recognized that ω depends on the local solute concentration [9] within the soil membrane, Eq.1 can still be used to determine a "global" osmotic coefficient even taking this dependency into account [10].

As soils and GCLs typically behave as nonideal or "leaky" membranes, diffusive flux does occur in response to the induced concentration gradient. Solute diffusion usually displays a transient phase, where the outflow concentrations change with time and a steady state, when the concentrations become constant. Although a more sophisticated pseudo-3D model [11] has been proposed for the interpretation of chemico-osmotic through-diffusion tests, in this study the traditional 1-D steady-state approach has been adopted, which consists in determining the cumulative mass per unit area, Q_t, of a given solute from the measurement of solute concentration in the outflow solutions [4]. The Q_t-versus-time plot usually shows a curved portion (transient diffusion) and a linear portion (steady-state diffusion). The intersection of the linear portion with the time axis is referred to as "time-lag", t_L. The slope of the linear portion (i.e., $\Delta Q_t/\Delta t$) is related to the chemo-osmotic diffusion coefficient, D_s^{ω}, as follows [4]:

$$D_s^{\omega} = H \frac{\left(\frac{\Delta Q_t}{\Delta t}\right)}{n(\bar{C}_t - \bar{C}_b)} \quad (3)$$

where $\bar{C}_t$= average solute concentration at top boundary, $\bar{C}_b$= average solute concentration at the base boundary, H = specimen thickness, and n = total soil porosity. In case of membrane behaviour, the chemo-osmotic diffusion coefficient can be expressed as [5]:

$$D_s^{\omega} = (1-\omega)\tau_m D_{so} \quad (4)$$

where τ_m is the matrix (geometric) tortuosity, D_{so} is the salt diffusion coefficient in free solution, and the product $\tau_m D_{so}$ is referred to as the effective diffusion coefficient, D_s^*. Assuming that solute sorption to soil is described by a linear equilibrium isotherm, t_L is related to the retardation factor R_d [-] as follows:

$$R_d = \frac{6\, t_L\, D_s^{\omega}}{H^2} \quad (5)$$

It should be noted that Eq.5 was derived under the assumption of a neutral solute undergoing linear sorption and "perfect flushing" boundary conditions

(i.e., constant solute concentration at the top and zero solute concentration at the base boundary).

Prior to the diffusion stage, the GCL specimens were first hydrated in flexible-wall permeameter (FWP) for 7 days; subsequently, permeation with DW was conducted for about 5 months under an average effective stress of 34.5 kPa to achieve saturation, remove soluble salts ("flushing") and to measure the reference k. An average head difference of 210 cm was maintained across the specimen during permeation.

After the flushing stage, the specimens were transferred to the rigid-wall cell and consolidated at two different effective vertical stress (70 kPa-Test 1 and 241 kPa-Test 2) by increasing the pressure in the upper chamber, to induce significantly different specimen heights and bulk void ratios. After the ensuing consolidation was complete, a further permeation stage with distilled water (DW) was conducted, mainly to check for sidewall leakage. The top piston was blocked against the steel frame to prevent volume changes of the specimen. Then, the chemo-osmotic diffusion test was initiated by circulating DW for 7 days at the specimen boundaries to establish a reference differential pressure (ΔP_{DW}), followed by circulation of the 20 mM KCl electrolyte solution at the top inflow line and DW at base inflow line. The circulation solutions at both boundaries of the specimen were periodically sampled every 2-3 days during the refill operations of the cylinders for pH and EC monitoring and for solute concentration analyses. The chemical analyses of the species contained in the inflow circulation solution (K^+, Cl^-) and of the main cations desorbed from the GCL (Na^+, Ca^{2+}) were carried out by a Dionex ICS 1000 Ionic Cromatograph.

2.3 Column test

A separate specimen (Test 3) was first subjected to the "flushing" procedure by means of permeation with DW, as described in the previous section. After about 5 months, permeation was temporarily stopped and the isotropic effective stress in the FWP was increased to 70 kPa, to simulate the stress level applied in Test 1. After the ensuing consolidation ended, the permeant liquid was switched from DW to 20 mM KCl solution and permeation was resumed. Bladder accumulators were used along both the inflow and outflow drainage lines; the effluent solutions were accumulated in the bladder and sampled over regular intervals for pH, EC and concentrations of chemicals species. The measured concentrations thus represent average incremental concentrations [12].

3 Results and discussion

3.1 Chemico-osmotic through-diffusion tests

3.1.1 Chemico-osmotic differential pressures

The different conditions of the specimens consolidated at different effective stresses were highlighted by the value of k to DW measured after consolidation, equal to $7.4 \cdot 10^{-11}$ m/s in Test 1 and $5.5 \cdot 10^{-12}$ m/s in Test 2, respectively. The differential pressure developed over time for the two specimens upon circulation with 20 mM KCl is shown in Fig.2. Test 1 developed a very small increase in differential pressure (ΔP=0.6 kPa) with respect to ΔP_{DW}, while for Test 2, ΔP increased by 3.1 kPa [8]. The corresponding values of the osmotic efficiency coefficient ω were 0.007 (Test 1) and 0.036 (Test 2), respectively. The results are qualitatively consistent with literature data, according to which the value of ω increases with the increase in consolidation stress applied prior to chemico-osmotic testing, and therefore with the reduction in the bulk void ratio, e_b. Nevertheless, the values of ω are significantly lower than those expected based on values reported for similar conventional GCLs and KCl concentration ($0.05 \leq \omega \leq 0.35$, [7]), presumably due to the quality of the bentonite lot, despite the GCL met the standard requirements for product acceptance (e.g., swell index, index flux, mass per unit area, etc.).

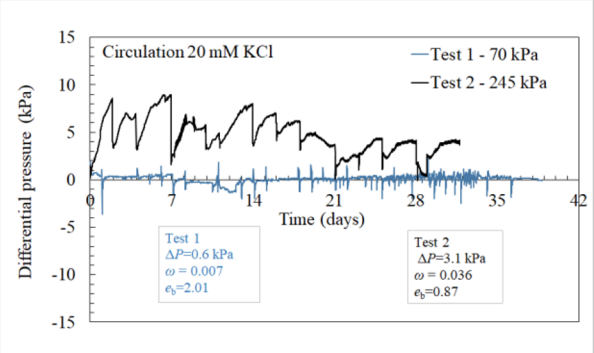


Figure 2 Chemico-osmotic differential pressure developed during circulation of 20 mM KCl solution in Test 1 and Test 2

3.1.2 Solute diffusion

Figure 3 shows the results of the chemical analyses conducted on the outflow solutions of Test 1 during circulation of 20 mM KCl, in terms of the trend of molar concentration over time of K^+, Cl^- and Ca^{2+}, which, unexpectedly, was the main desorbed cation from the GCL during circulation of KCl at both boundaries of the specimen, whereas Na^+ was mainly eluted from the GCL during permeation and circulation with DW. Figure 4 shows the results of the chemical analyses conducted on the outflow solutions of Test 2 during circulation of 20 mM KCl, in terms of the trend of molar concentrations over time of K^+, Cl^- and Na^+, which, was the main cation eluted from the GCL, also during the flushing stage and preliminary circulation with DW. Although chemical characterization of the bentonite (section 2.1) showed the dominance of Na^+ over Ca^{2+} in the exchange complex and Ca^{2+} should be retained more tightly than Na^+ onto surface sites upon cation exchange reactions based on valence, the observed dominance of Ca^{2+} with respect to Na^+ in the outflow solutions of Test 1 probably reflects significant variability in the distribution of adsorbed cations of the bentonite with the same GCL roll.

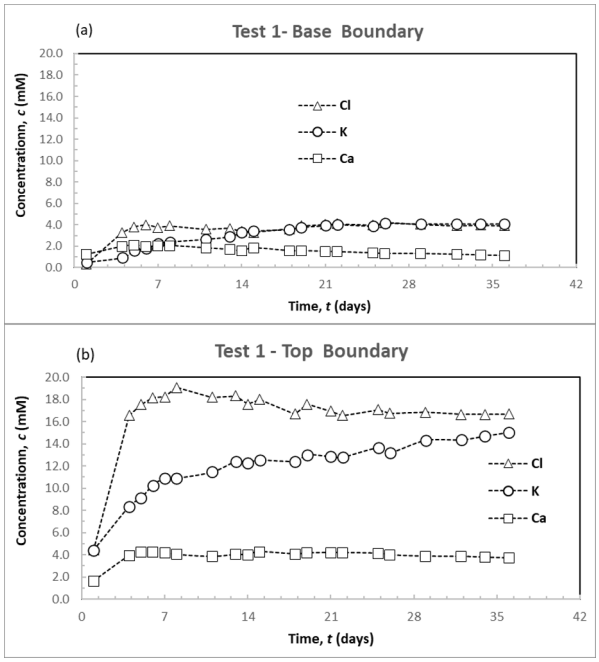


Figure 3. Test 1 - Concentration of major anions and cations in the outflow solutions at base (a) and top (b) boundary during circulation of 20 mM KCl as source solution

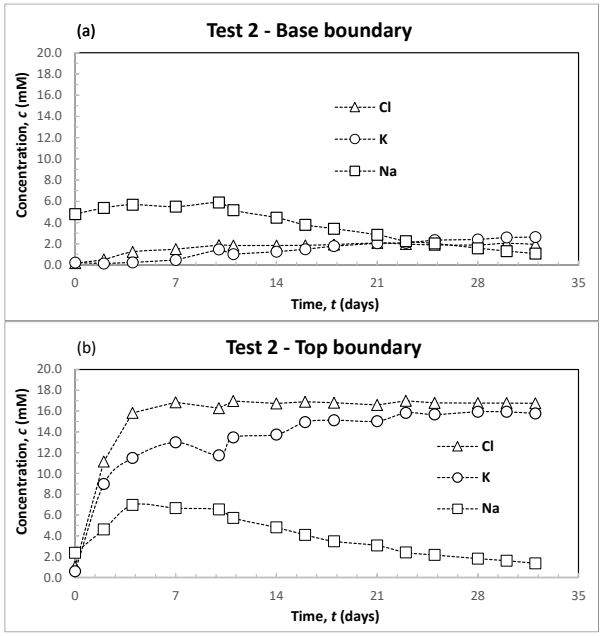


Figure 4. Test 2 - Concentration of major anions and cations in the outflow solutions at base (a) and top (b) boundary during circulation of 20 mM KCl as source solution

3.1.3 Solute transport parameters

The Q_t versus time plots for both Cl^- and K^+ are shown in Fig.5 and Fig 6 for Test 1 and Test 2, respectively. The assessments of the time lags, t_L, by extrapolation of the linear part of the plots to the intersection with the time axis, are also shown in Fig. 5 and Fig. 6. The slopes of the linear part of the plots were used in Eq.3 to calculate the diffusion coefficients and Eq.5 was then used to calculate the retardation factors R_d of K^+ and Cl^- in each test. The calculated solute transport parameters are reported in Table 1.

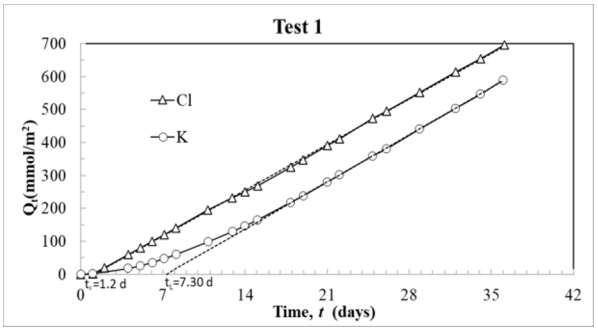


Figure 5. Test 1 – Cumulative masses of Cl^- and K^+ per unit area and assessment of the time lag t_L

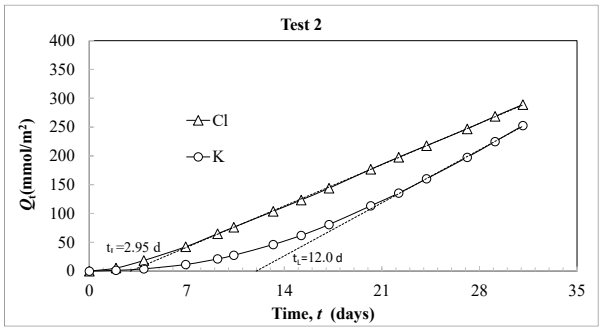


Figure 6. Test 2 – Cumulative masses of Cl^- and K^+ per unit area and assessment of the time lag t_L.

Table 1. Solute transport parameters from diffusion tests.

Test	H (mm)	n (-)	ω (-)	Ion -	D_s^{ω} ($\cdot 10^{10}$ m²/s)	t_L (days)	R_d (-)	K_d (cm³/g)
1	5.8	0.67	0.007	Cl^-	1.23	1.2	2.2	1.2
				K^+	1.35	7.3	15.0	13.8
2	4.0	0.45	0.036	Cl^-	0.61	2.95	5.9	2.1
				K^+	0.84	12.0	33.3	13.7

For both tests, given the low values of ω, the determined values of D_s^{ω} can be assumed to practically coincide with the effective diffusion coefficient D_s^*, that only accounts for the matrix tortuosity. Moreover, the diffusion coefficients determined for the anion and the cation species are very similar, in accordance with the electroneutrality constraint. As expected, the diffusion coefficients are lower for the GCL specimen consolidated to higher stress, owing to the reduction in porosity and matrix tortuosity factor.

Based on the calculated R_d, both the anion and cation species were adsorbed to the GCL. Moreover, the R_d for both K^+ and Cl^- were higher at higher stress, i.e. at lower void ratio, consistently with the definition of R_d, which can be also expressed as:

$$R_d = 1 + \frac{\rho_s K_d}{e} \quad (6)$$

where ρ_s= density of solids, K_d= distribution coefficient for linear adsorption, e= soil void ratio. Note that to calculate K_d, the density of bentonite solids

(ρ_s=2.65 g/cm^3), and the bulk void ratio of the GCL specimen were used in Eq.6.

Retardation of K^+ can be mainly ascribed to electrostatic attraction onto negatively charged clay solid surface following exchange with either Na^+ or Ca^{2+}, which were desorbed into the circulation solutions. In any case, the calculated distribution coefficients of K^+ are practically the same for both tests. Retardation factors of Cl^- (usually assumed as a conservative tracer) greater than one have previously been reported from diffusion tests on GCLs (e.g., [4],[12-13]). The retardation of Cl^-, consistently inferred by both test results, can be ascribed to some anion adsorption capacity at clay particle edges (e.g., due to broken bonds), as well as to accuracy of the chemical analyses.

3.2 Column test

3.2.1 Hydraulic conductivity and solute breakthrough curves

The hydraulic conductivity, *k*, of the GCL specimen during permeation with the 20 mM KCl solution is plotted in Fig.7 (left axis) versus the number of pore volumes of flow, *PVF*. The permeation stage lasted 238 days, corresponding to *PVF*=15.8. Fig. 7 also shows (left axis) the solute breakthrough curves of Cl^- and K^+, in terms of the relative effluent concentration (i.e., *C*/*C*o, where *C*o is the influent concentration). The *k* remained practically constant during permeation, suggesting the absence of significant adverse effects of the permeant solution on the permeability of bentonite, at least within the duration of the test. It can also be observed that Cl^- reached complete breakthrough (i.e., *C*/*C*o=1) whereas K^+, due to greater retardation, barely reached *C*/*C*o=0.5. Na^+ and Ca^{2+} cations were mainly desorbed into the effluent solutions, due to exchange with K^+ cations .

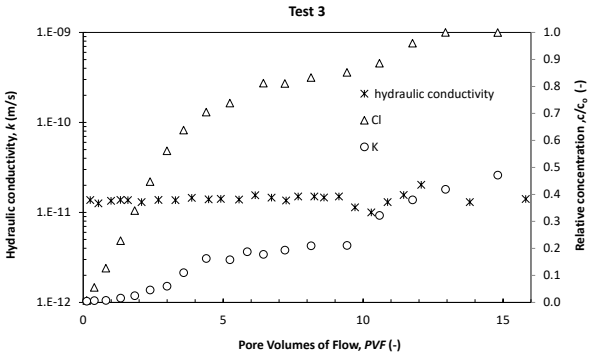


Figure 7. Test 3 –Hydraulic conductivity versus the *PVF* during permeation with 20 mM KCl solutions (left axis) and solute breakthrough curves of Cl^- and K^+(right axis). *C*/*C*o values are plotted in the middle of the sampling interval.

3.2.2 Solute transport parameters

Considering that *k* and the average hydraulic gradient (*i*=290) were practically constant throughout the test, the seepage velocity, *v*, can also be considered constant (v=6.1·10^{-9} m/s). Moreover, since no significant membrane behaviour was observed in Test 1, that was conducted identically to Test 3 in terms of flushing stage and applied consolidation stress, it can be assumed that no significant membrane effects occurred during the column test. Therefore, the solute migration analysis of the column test can be carried out by the standard ADRE approach [12].

[14] recommended that, when column tests are conducted using 'bladder' type accumulators (as in the present study) the "dilution" effect induced by the volume of the accumulator is taken into account. Therefore, the solute curves of K^+ and Cl^- have been regressed with the above-mentioned theoretical approach by quantifying the "dead" volume V_d, of the used apparatus (35 mL). In Fig.8 the best-fit theoretical breakthrough curves are compared with the experimental curves and the resulting values of the transport parameters for Cl^- and K^+ are reported in Tab. 2 together with the corresponding parameters obtained for Test 1.

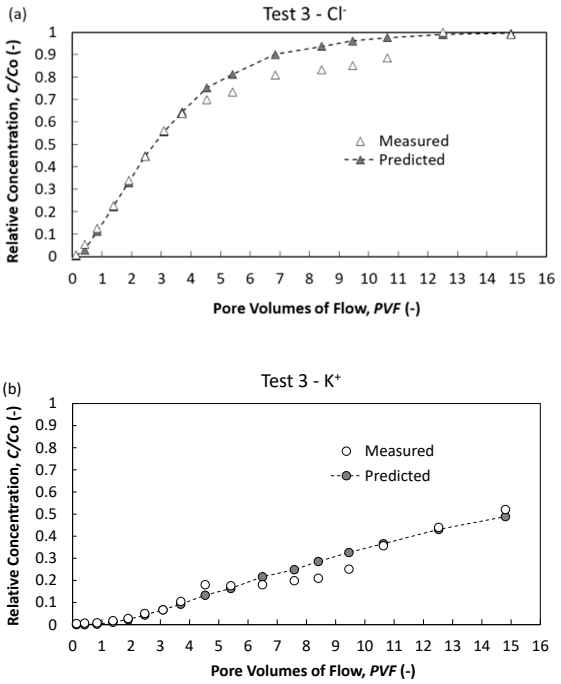


Figure 8. Test 3 –Measured and best-fit theoretical solute breakthrough curves of Cl^- and K^+.

Table2. Solute transport parameters from diffusion and column tests.

Test	*H* (mm)	*n* (-)	*ω* (-)	Ion -	D_s^{ω}/D_h ($\cdot 10^{10}$m^2/s)	t_L (days)	R_d (-)	K_d (cm^3/g)
1	5.8	0.67	0.007	Cl^-	1.23	1.2	2.2	1.2
				K^+	1.35	7.3	15.0	13.8
3	7.2	0.68	-	Cl^-	1.35	-	2.5	1.6
				K^+	1.42	-	20.0	19.8

The dispersion coefficients, D_h, estimated for K^+ and Cl^- were very similar (Test 3), whereas the estimated R_d for K^+ was significantly higher than for Cl^-, consistently with the result of the through-diffusion test. Given the low seepage velocity and small thickness of the specimen (Péclet number, $Pe \cong 0.3$), mechanical dispersion is negligible, such that the inferred dispersion coefficients for K^+ and Cl^- can be considered effective diffusion coefficients (D^*), in analogy with the results of through-diffusion tests. The results in Tab.2 show that the effective diffusion coefficients obtained by the two testing approaches were very similar. The estimated R_d for Cl^-(2.5) was also very close to that obtained in the diffusion test (2.2), confirming the small retardation of Cl^-. The estimated R_d and K_d of K^+ were higher in the column test, which could depend on the longer test duration. In essence, considering also the negligible impact of membrane behaviour of the GCL used in this study, the two testing approaches provided similar results of transport parameters of K^+ and Cl^-.

4 Conclusions

Chemico-osmotic through-diffusion tests were performed using a 20 mM KCl solution on GCL specimens previously consolidated at two effective stresses (70 kPa and 241 kPa). The measured values of the osmotic efficiency coefficient (ω=0.007 and ω=0.0369, respectively), although lower than expected based on the literature, qualitatively confirmed the increase in ω with the consolidation stress and thus with the decrease in the GCL bulk void ratio. The diffusion coefficients D_s^* and R_d were respectively lower and higher at higher stress, owing to the reduction in matrix tortuosity factor and porosity. A column test was performed at effective stress of 70 kPa, using a 20 mM KCl solution and provided solute transport parameters close to those obtained in the through-diffusion test. The results confirmed the enhancing effect of consolidation on solute containment properties of GCLs, owing to the decrease in effective diffusion coefficients and the concurrent increase in chemico-osmotic efficiency and in retardation factors.

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