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Theoretical assessment of the advective-diffusive transport of contaminants through landfill composite liners

Évaluation théorique du transport advectif-diffusif de contaminants permis les barrières composites des décharges

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ABSTRACT: Composite liners consisting of a geomembrane, placed over a low-permeability clay layer, are used throughout the world for the lining of waste disposal facilities, with the aim to prevent the groundwater quality from being compromised, and an unacceptable risk for the human health and the environment from occurring. Despite the importance of developing a rational approach to calculate the contaminant transport rate through composite liners, for both the design of new landfills and the risk assessment of the existing ones, limited attention has been paid so far to the derivation of analytical, numerical, or hybrid analytical-numerical solutions, which can help designers assess the containment performance of lining systems. After an overview of the simplified calculation approaches that are currently adopted for the performance-based design of composite liners, a novel theoretical framework is presented in this paper, which allows the advective-diffusive transport of inorganic contaminants to be modelled considering imperfect contact conditions between the geomembrane and the underlying clay layer.

RÉSUMÉ: Les barrières composites constituées d'une géomembrane, placée sur une couche d'argile à faible perméabilité, sont utilisés dans le monde pour l'isolement des installations de stockage des déchets, pour éviter que la qualité des eaux souterraines ne soit pas compromise et ne présente pas un risque inacceptable pour la santé et l'environnement. Malgré l'importance de développer une approche rationnelle pour calculer le taux de transport des contaminants permis les barrières composites, pour la conception de nouvelles décharges et pour l'évaluation des risques des décharges existantes, une attention limitée a été accordée jusqu'à présent à l'élaboration de solutions analytiques, numériques ou hybrides analytiques-numériques, qui peuvent aider les ingénieurs à évaluer les performances de confinement des systèmes d'isolement. Après un aperçu des approches de calcul simplifiées actuellement adoptées pour la conception basée sur les performances des barrières composites, un nouveau cadre théorique est présenté dans cet article, qui permet de modéliser le transport advectif-diffusif des contaminants inorganiques en considérant des conditions de contact imparfaites entre la géomembrane et la couche d'argile sous-jacente.

Keywords: Contaminant transport; geosynthetics; landfills; performance-based design; risk assessment.

1 INTRODUCTION

Composite liners that include a high-density polyethylene (HDPE) geomembrane (GM), placed over a compacted clay liner (CCL), are prescribed throughout the world for the bottom lining systems of waste disposal facilities, with the goal of minimising the migration of contaminants from the waste fill to the surrounding environment (Giroud and Cazzuffi, 1989). Furthermore, the use of alternative lining systems, such as those in which the CCL is replaced with a geosynthetic clay liner (GCL), is permitted in most regulations, provided that equivalency with the prescribed composite liner is demonstrated on the basis of a selected performance criterion. The most widely adopted approach is to assume that equivalency is demonstrated if the steady-state advective travel time through the proposed alternative liner is lower than that through the prescribed liner, but this approach fails to recognise that diffusion is a significant, if not the dominant, contaminant transport mechanism within low-permeability clay liners (Manassero and Shackelford, 1994; Foose et al., 2002; Shackelford, 2014). For such a reason, the performance criteria that account for the combined effect of advection and diffusion on contaminant transport, under either steady-state or transient-state conditions, are preferable over the advective travel time criterion (Shackelford, 1990; Manassero et al., 2000; Katsumi et al., 2001; Rowe and Brachman, 2004).

Although several analytical, numerical, and empirical solutions have been developed, since the pioneering study of Giroud and Bonaparte (1989), to calculate the liquid flux through landfill composite liners, relatively few studies have been aimed at quantifying the advective-diffusive transport of contaminants in the presence of defects in the GM layer (e.g., defective seams between adjacent panels, punctures caused by sharp materials beneath and above the GM, and tensile failures induced by the landfilling operations). The latter issue is particularly relevant in the case of inorganic compounds, which, unlike many organic compounds, do not readily diffuse through intact portions of the GM and, therefore, their transport should be recognised as a three-dimensional (3D) process that involves migration through the GM defects, through the interfacial zone between the GM and the underlying low-permeability mineral layer and, finally, through the mineral layer itself, via a combination of advection and diffusion.

A simplified approach to model the advective-diffusive transport of inorganic contaminants through composite liners was proposed by Katsumi et al. (2001), who identified an equivalent one-dimensional (1D) system for which analytical solutions to the contaminant breakthrough time and mass flux exist. This approach, which has since been implemented in a number of analytical studies that have dealt with the performance-based design of landfill lining systems, allows both steady-state (Foose, 2010; Guarena et al., 2020; Dominijanni and Manassero, 2021; Dominijanni et al., 2021) and transient-state contaminant transport analyses (Foose et al., 2001; Kandris and Pantazidou, 2012; Dominijanni et al., 2023) to be carried out using typical spreadsheet applications and hand-held calculators. Despite being particularly attractive because of its versatility, the Katsumi et al. (2001) approach is not devoid of drawbacks, the most serious of which can probably be ascribed to neglecting the mass conservation condition for the solute phase. The objective of this paper is thus to illustrate a novel theoretical framework, which has recently been developed by Guarena et al. (2023) with the aim of covering the aforementioned modelling gap.

2 THEORETICAL FRAMEWORK

The theoretical framework outlined herein has been developed under the assumption that inorganic contaminants preferentially migrate through holes that are located in correspondence to the wrinkles of the GM layer, rather than through holes that occur in flat areas. Therefore, only the case of a damaged wrinkle of width $2b_w$ is referred to in this paper, which is perfectly analogous to the case of a cut, tear, or defective seam of width $2b_w$ occurring in a flat area of the GM if the liquid and contaminant transport rates are not controlled by the size of the actual holes in the wrinkle (Figure 1). Moreover, if the length of the defect, L_w, is much greater than its width (i.e., defect of uniform width and infinite length), the liquid and contaminant transport analyses can be treated as two-dimensional problems (Touze-Foltz et al., 1999; Giroud and Touze-Foltz, 2005).

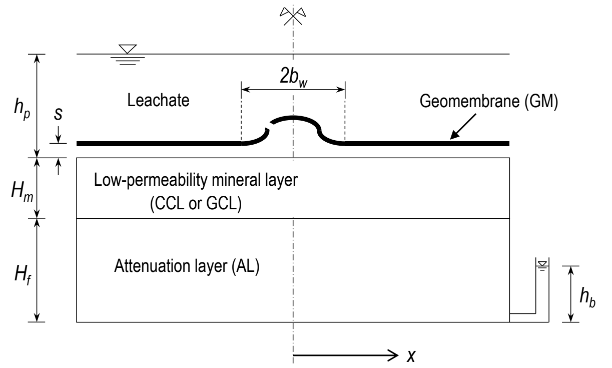

Figure 1. Reference scheme for the liquid and contaminant transport analyses through a composite liner with a defect of uniform width and infinite length in the GM layer and imperfect contact conditions between the GM and the low-permeability mineral layer (not to scale).

2.1 Katsumi et al. (2001) approach

The approach that was proposed by Katsumi et al. (2001), with the aim of providing a simple calculation tool to analyse the transport of inorganic contaminants through composite liners when the leachate flow is at steady state, consists in computing an equivalent area, A_e, which conducts the same (total) leachate flow rate as the considered defect for the same hydraulic head drop across the composite liner. If Rowe's (1998) analytical model for non-interacting defects of uniform width and infinite length is adopted to assess the leachate flow rate, the equivalent area is expressed as follows:

$$A_e = 2b_w L_w \left(1+\frac{1}{\alpha b_w}\right) \tag{1}$$

The α parameter that appears in Equation 1 can be interpreted as the exponential decay constant of the hydraulic head beneath the GM, and is given by:

$$\alpha = \sqrt{\frac{k_s}{\left(H_f + H_m\right)\theta_h}} \tag{2}$$

where θ_h is the hydraulic transmissivity of the interfacial zone between the GM and the low-permeability mineral layer, and k_s is the equivalent hydraulic conductivity corresponding to the low-permeability mineral layer (saturated hydraulic conductivity k_m) and the attenuation layer (AL) (saturated hydraulic conductivity k_f):

$$k_s = \frac{H_f + H_m}{\dfrac{H_f}{k_f} + \dfrac{H_m}{k_m}} \tag{3}$$

Once the equivalent area has been computed, the contaminant mass flow rate through the GM defect, J_e, can be obtained as the product of A_e and the contaminant mass flux, which in turn is calculated according to the existing analytical solutions to the partial differential equation that governs the 1D solute transport through multi-layerd barriers via advection, diffusion, sorption, and degradation. When steady-state conditions are achieved for both the liquid and solute transport, the dimensionless contaminant mass flow rate through the GM defect, J_e^*, which is defined as the ratio of J_e to the advective component of the mass flow rate occurring below the defect, is given by:

$$J_e^* = \left(1 + \frac{1}{\alpha b_w}\right)\frac{\exp(P_L) - \chi_b}{\exp(P_L) - 1} \tag{4}$$

where P_L is the Péclet number and χ_b is the ratio of the contaminant concentration at the bottom of the composite liner to that in the leachate drainage layer.

The Péclet number can be assessed as follows:

$$P_L = \frac{i_s k_s}{\Lambda} \tag{5}$$

where i_s is the maximum mean hydraulic gradient through the low-permeability mineral layer and the attenuation layer, and Λ is the equivalent diffusivity:

$$i_s = 1 + \frac{h_p - h_b}{H_f + H_m} \tag{6}$$

$$\Lambda = \frac{1}{\dfrac{H_f}{n_f D_f^*} + \dfrac{H_m}{n_m D_m^*}} \tag{7}$$

being n_m and n_f the porosities of the low-permeability mineral layer and the attenuation layer, respectively, and D_m^* and D_f^* the effective diffusion coefficients of the low-permeability mineral layer and the attenuation layer, respectively, which are obtained as the product of the apparent tortuosity factor, τ (< 1), and the free-solution diffusion coefficient of the contaminant, $D_{s,0}$.

2.2 Guarena et al. (2023) approach

According to the theoretical framework which has recently been developed by Guarena et al. (2023), the 3D advective-diffusive transport of inorganic contaminants within the composite liner is conceptualised as a horizontal flow along the interface between the GM and the low-permeability mineral layer and then a vertical flow in the low-permeability mineral layer, in a similar way to the reference scheme that was considered by Rowe (1998) and Touze-Foltz et al. (1999, 2001) with a view to approximating the actual leachate flow network under imperfect contact conditions at the GM/CCL or GM/GCL interface.

If steady-state conditions are achieved for both the liquid and solute transport, the contaminant concentration profile beneath the GM satisfies the following homogeneous, second-order, linear ordinary differential equation with non-constant coefficients:

$$\begin{aligned}&\frac{\mathrm{d}^2\chi}{\mathrm{d}\xi^2} - \frac{\Theta}{\exp(\xi)}\frac{\mathrm{d}\chi}{\mathrm{d}\xi} + \\ &- \frac{\Theta}{\exp(\xi)\left[\exp\left(\dfrac{P_L}{\exp(\xi)}\right) - 1\right]}(\chi - \chi_b) = 0\end{aligned} \tag{8}$$

which has to be solved for $0 \le \xi < +\infty$ in conjunction with the following set of boundary conditions:

$$\begin{cases} \chi(0) = 1 \\ \lim\limits_{\xi \to +\infty} \dfrac{\mathrm{d}\chi}{\mathrm{d}\xi}(\xi) = 0 \end{cases} \tag{9}$$

In Equation 8, χ is the ratio of the contaminant concentration beneath the GM to that in the leachate drainage layer, ξ is the dimensionless distance from the GM defect, $\xi = \alpha(x - b_w)$, and Θ is the transmissivity number, which is a measure of the relative importance of advection compared to diffusion through the GM/CCL or GM/GCL interface:

$$\Theta = \frac{i_s \theta_h (H_f + H_m)}{\theta_d} \tag{10}$$

where θ_d the diffusive transmissivity of the interface.

Once the steady-state contaminant concentration profile beneath the GM has been determined, the dimensionless contaminant mass flow rate through the GM defect, J_s^*, is given by:

$$J_s^* = \frac{\exp\left(P_L\right) - \chi_b}{\exp\left(P_L\right) - 1} + \frac{1}{\alpha b_w}\left[1 - \frac{1}{\Theta}\frac{\mathrm{d}\chi}{\mathrm{d}\xi}\left(\xi = 0\right)\right] \quad (11)$$

3 EXAMPLE ANALYSIS

With the aim of quantifying the error in the predicted steady-state contaminant mass flow rate, due to the use of the Katsumi et al. (2001) approach relative to the novel theoretical framework outlined in this paper, and referred to as the Guarena et al. (2023) approach, the lining system described in Table 1 has been considered herein. Such lining system comprises, from top to bottom, a GM, a 1-cm-thick GCL, and a 4-m-thick AL. Cadmium (Cd^{2+}) has been selected to represent the inorganic leachate constituent of interest ($D_{s,0}$ = $7.17 \cdot 10^{-10}$ m^2/s), and the half-width of the GM defect, b_w, has been assumed equal to 0.1 m. The GM-GCL interface has been assigned a value of the hydraulic transmissivity ($\theta_h = 3.5 \cdot 10^{-11}$ m^2/s), which is consistent with the range of variation measured through laboratory-scale experiments (Rowe and Brachman, 2004).

Table 1. Properties of the layers comprising the landfill composite liner, which is referred to for the purposes of the example analysis.

	AL	**GCL**	
n	0.3	0.69	(-)
τ	0.25	0.31	(-)
k	$1 \cdot 10^{-7}$	$1 \cdot 10^{-11}$	(m/s)

Perfect flushing boundary conditions have been adopted at the base of the composite liner (i.e., $\chi_b = 0$), while the diffusive transmissivity of the GM-GCL interface, in the absence of experimental studies devoted to its measurement, can be tentatively estimated as follows:

$$\theta_d = D_{s,0} s \quad (12)$$

where s is the thickness of the GM/GCL interface, which in turn can be related to θ_h, if Newton's viscosity law for the flow between two parallel plates applies to the transmissive layer (Giroud and Bonaparte, 1989):

$$s = \sqrt[3]{\frac{12\theta_h \mu_w}{\gamma_w}} \quad (13)$$

being γ_w (10 kN/m^3) and μ_w (10^{-6} kPa·s) the unit weight and viscosity of water, respectively.

According to Equation 12, the θ_d parameter varies between $1 \cdot 10^{-15}$ and $1 \cdot 10^{-14}$ m^3/s, and the associated range of variation of the transmissivity number can be assessed through Equation 10 as a function of the hydraulic gradient. As shown in Figure 2a, even when diffusion represents the dominant transport mechanism within the multi-layered soil profile (i.e., $P_L \ll 1$), contaminant transport through the GM/GCL interface may be controlled by advection due to the high values of the transmissivity number (i.e., $\Theta \gg 1$) corresponding to the estimated values of the diffusive transmissivity.

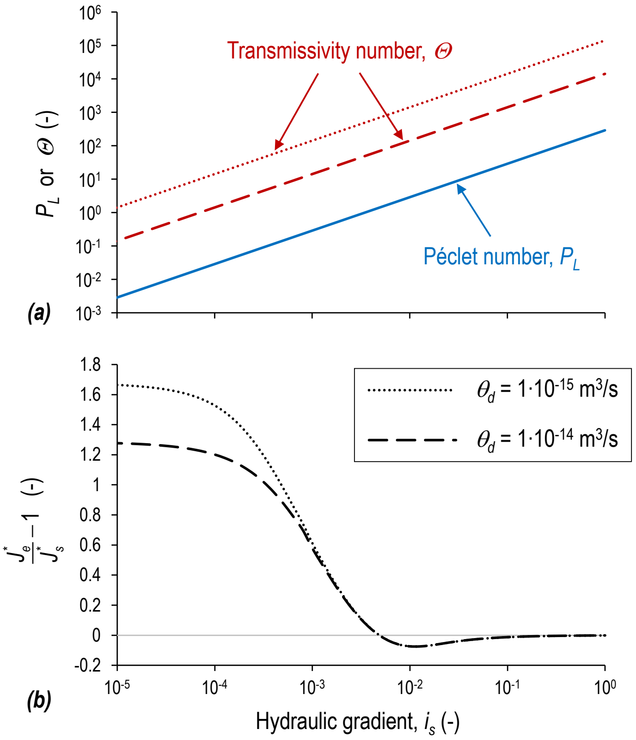

Figure 2. Results of the example calculation: (a) Péclet and transmissivity numbers as a function of the hydraulic gradient across the composite liner, and (b) error in the predicted contaminant mass flow rate due to the use of the Katsumi et al. (2001) approach.

The error in the predicted steady-state contaminant mass flow rate, which is associated with the use of the Katsumi et al. (2001) approach, can be finally assessed for the considered range of variation of both the hydraulic gradient and the diffusive transmissivity. Equations 4 and 11 have been used to calculate J_e^* and J_s^*, respectively, the latter of which has been determined after solving numerically Equation 8 according to the finite difference method.

As shown in Figure 2b, two asymptotic conditions clearly emerge from the example analysis:

- at the highest hydraulic gradients ($i_s > 10^{-2}$), when advection dominates over diffusion through both the multi-layered soil profile ($P_L \gg 1$) and the GM/GCL interface ($\Theta \gg 1$), the Katsumi et al. (2001) approach leads to a prediction of the contaminant mass flow rate that can be regarded as the rigorous one from the mass conservation condition viewpoint, for both the liquid and solute phases;
- at the lowest hydraulic gradients ($i_s < 10^{-3}$), when the importance of diffusion is comparable to ($\Theta \sim 1$) or greater than ($P_L \ll 1$) that of advection through the GM/GCL interface and the multi-layered soil profile, respectively, neglecting the mass conservation condition for the solute phase causes an error, whose magnitude depends to a large extent on the diffusive transmissivity of the GM/GCL interface.

4 CONCLUSIONS

An original theoretical framework has recently been developed by Guarena et al. (2023) to calculate the steady-state mass flow rate of inorganic contaminants through landfill composite liners, which are made up of a GM overlying a low-permeability mineral layer. The transport of both the liquid and the solute has been assumed to only occur through GM defects of uniform width and infinite length (e.g., holed wrinkles and defective seams), with the additional restriction that all the soil layers maintain fully-saturated conditions. The migration pathway within the lining system has been conceptualised as a horizontal flow along the interfacial zone between the GM and the underlying mineral layer, and then as a vertical flow in the mineral layer itself, as in the reference scheme considered by Rowe (1998) and Touze-Foltz et al. (1999, 2001) for the calculation of the leachate flow rate. The theoretical framework developed by Guarena et al. (2023) can thus be interpreted as an attempt to take a step forward with respect to the simplified calculation approach, which was originally conceived by Katsumi et al. (2001) and is routinely being used for the performance-based design of landfill lining systems.

The error associated with the use of the simplified calculation approach proposed by Katsumi et al. (2001) has been quantified in the present paper with the aid of an example analysis. In the case of purely advective transport, the latter approach has been proven to be consistent with the mass conservation condition for both the solvent and solute phases, while satisfaction of the mass conservation condition for the solute phase is no longer guaranteed when diffusion represents the main transport mechanism, leading to an error that greatly depends on the diffusive properties of the interface between the GM and the mineral layer.

Further research is recommended on the aspects dealt with in the present paper. From a theoretical viewpoint, solutions to the contaminant mass flow rate should be developed for shapes of the geomembrane defects (e.g., circular holes in a flat GM), boundary conditions (e.g., parallel interacting damaged wrinkles), transport properties of the soil layers (e.g., change in the degree of saturation for positive suction heads), and chemical types (e.g., organic contaminants diffusing through intact GMs) that differ from the working hypotheses of this study. From an experimental viewpoint, the laboratory investigation of the relative role of advection and diffusion in the transport of contaminants through the GM/CCL and GM/GCL interfaces is of the utmost importance. Finally, it would be advisable to validate the outlined theoretical framework through comparison of the model predictions with actual field measurements, e.g., the contaminant concentration in the secondary leachate collection system of a double composite liner.

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