

Transport of VOCs through a Co-extruded Geomembrane with a Nylon Barrier

R.S. McWatters and R.K. Rowe, GeoEngineering Centre at Queen's-RMC, Department of Civil Engineering, Queen's University, Kingston, Ontario, Canada.

ABSTRACT

Geomembranes are employed in a variety of containment and barrier systems to mitigate the migration of hazardous volatile organic compounds (VOCs) with the potential to contaminate the surrounding environment and air quality. In addition, they are often the primary barriers used in final cover systems of landfills to prevent the release of these contaminants found in leachate and landfill gas. Studying the transport of VOCs through a geomembrane establishes its diffusive and permeation properties and ultimately its performance as a barrier. This paper examines the liquid and gas phase transport of VOCs through a novel co-extruded linear low density polyethylene geomembrane with an inner nylon layer. The 0.38 mm (15 mil) nylon geomembrane showed a 5-15 fold decrease in both the permeation and diffusion coefficients when compared to a standard 0.53 mm (20 mil) LLDPE geomembrane.

1. INTRODUCTION

Geomembranes have been used in the engineering industry for a variety of applications, including containment of waste in landfills and disposal facilities, for surface ponds or reservoirs, and containment basins for fuel and liquid storage. More recently, geomembrane applications have extended to include vapour barriers for buildings, whether as a methane barrier or to prevent harmful vapours from entering through the building foundation. As a vapour barrier, geomembranes also contain landfill gases including, but not limited to, methane, carbon dioxide and volatile organic compounds (VOCs) (Qian *et al.*, 2001). In addition, they have been employed in the remediation of soil piles contaminated by hydrocarbons and to contain gases released during the degradation process in bioreactors (Reinhart *et al.*, 2002; Rowe *et al.*, 2004). As the geotechnical and geoenvironmental industries continue to develop more uses for geomembranes, the types of geomembranes available for use have also grown.

Common polymer types used in barrier systems include high density polyethylene (HDPE), linear low density polyethylene (LLDPE), poly-vinyl chloride (PVC), and polyethylene liners with fluorinated additives (Park and Nibras, 1993). Multilayer geomembranes were first created having higher density polyethylene as the outside layers and a lower density polyethylene as the inner layer (Kolbasuk, 1990). Evolved from multi-layer geomembranes were co-extruded geomembranes with additives, as in a polyamide (nylon) material as the innermost layer. The objective was to create a geomembrane with the flexibility and stability of a standard LLDPE polymer but with a superior resistance to vapour and liquid VOC migration. This paper examines the liquid and gas phase diffusion of VOCs through a novel co-extruded linear low density polyethylene geomembrane with an inner nylon barrier.

2. BACKGROUND

As an effective barrier material in landfill cover systems or hazardous containment systems, geomembranes must be resistant to the migration of VOCs found in landfill gas. For a well constructed and intact geomembrane, there is minimal advective transport of VOCs and diffusion is the dominant mode of transport (Rowe, 1998). The diffusive flux through the geomembrane will depend on polymer type and crystallinity. Crystallinity and polymer orientation have been shown to impact permeability as a tighter packing structure results in a more tortuous path for compounds to navigate, thus reducing the permeability (D'Aniello *et al.* 2000; Aminabhavi *et al.*, 1997). Geomembrane thickness is another important factor as thinner geomembranes have a higher diffusive flux, other things being equal (Sangam, 2001). A polyamide material used in a co-extruded geomembrane could increase the diffusive resistance to VOCs for a thin geomembranes.

Three parameters characterize the potential for diffusive migration of each compound through a geomembrane: diffusion, partitioning and permeation coefficients (Sangam and Rowe, 2001). Diffusion through geomembranes occurs in three steps: adsorption, diffusion and desorption. Initially, the contaminant partitions between the source medium and adjacent surface of the geomembrane. Then the compound diffuses through the geomembrane driven by chemical potential. Finally, the compound partitions between the outer geomembrane surface and the receiving medium (Sangam and Rowe, 2001). Eventually, equilibrium is reached between the concentration in the geomembrane and the

concentration in either the source or receptor media (Rowe, 1998). The equilibrium between geomembrane and the source medium can be related by Henry's law:

$$c_g = S_{gf} c_f, \quad (1)$$

where c_g is the concentration in the geomembrane [ML^{-3}], c_f is the concentration in the source fluid (either gas or liquid) [ML^{-3}], and S_{gf} is the partitioning coefficient [-].

In the second step, the diffusion of the penetrant through the geomembrane is described by Fick's first law:

$$f = -D_g \frac{dc_g}{dz}, \quad (2)$$

where f is the mass flux [$\text{ML}^{-2}\text{T}^{-1}$]. The diffusion coefficient, D_g [L^2T^{-1}] is specific to the geomembrane and contaminant of interest. c_g is the concentration of the compound in the geomembrane [ML^{-3}] and z represents the distance parallel to the direction of transport. When the diffusion coefficient is constant, the change in penetrant concentration in the geomembrane with time t , is expressed by Fick's second law:

$$\frac{\partial c_g}{\partial t} = D_g \frac{\partial^2 c_g}{\partial z^2} \quad (3)$$

The final step is similar to the first and can also be described by Henry's law with different notation :

$$c'_g = S'_{gf} c_f, \quad (4)$$

where c'_g is contaminant concentration in the receptor medium and S'_{gf} is the partitioning out of the geomembrane. When the source and receptor fluid are the same, the partitioning coefficients into the geomembrane can be assumed to be equal to the partitioning coefficient out of the geomembrane ($S_{gf}=S'_{gf}$) (Sangam and Rowe, 2001).

The concentration of contaminant inside the geomembrane is very difficult to measure; therefore, the change in concentration in the fluid on either side of the geomembrane is measured. The relationship for this change in concentration in the geomembrane when the source and receptor media are alike is given by rearranging Equations 1 and 3. The relationship is as follows:

$$f = -D_g \frac{dc_g}{dz} = -S_{gf} D_g \frac{dc_f}{dz}, \quad (5)$$

Further simplification of this equation in terms of a permeation coefficient, P_g [L^2T^{-1}], which is the coefficient of proportionality for Fickian mass transport for a source to receptor fluid (Sangam and Rowe, 2001), is shown below:

$$P_g = S_{gf} D_g \quad (6)$$

The diffusive properties of a geomembrane (S_{gf} , D_g , P_g) with respect to VOC contaminants in a liquid or vapour phase are established through sorption and diffusion tests. Lower permeation coefficients are indicative of a more effective barrier to the contaminant being investigated.

3. EXPERIMENTAL INVESTIGATIONS

3.1 Material and Methods

This study examined a relatively new co-extruded flexible geomembrane supplied by Raven Industries, Engineered Films Division (Sioux Falls, South Dakota, USA). The geomembrane is marketed as a nylon vapour barrier, trade name VBP15. It is 0.38 mm (15 mil) thick with a five layer structure. The first layer is approximately 42% of the total thickness

and made of low linear density polyethylene (LLDPE) with a 3.5% calcium carbonate inert filler. The second layer, at 6% of the total thickness, is a tie resin made of maleic anhydride modified LLDPE. The innermost layer is only 4% of the thickness and made of nylon. The fourth and fifth layers are identical to the first and second respectively, therefore creating a symmetrically layered geomembrane. The density of the geomembrane is 920 kg/m³. The coextruded geomembrane was compared with a standard 0.53 mm (20 mil) LLDPE geomembrane also manufactured by Raven Industries. The density of this geomembrane was 920 kg/m³. Table 1 and 2 summarize the relevant properties of the co-extruded and standard geomembrane tested, respectively. All properties were obtained from the manufacturer.

Table 1. Standard properties of co-extruded LLDPE geomembrane (with an inner nylon barrier) tested.

Properties	Methods (ASTM)	Units	Values
Thickness		mm	0.38
Puncture	D4833	Peak (N)	171.52
Tear resistance	D1004	Peak (N)	39.46 x 39.32
Tensile	D6693	Peak (N)	72.73
Load at break	D6693	N	70.15
Elongation at break	D6693	% (min)	608
Tensile strength	D882	N	298
Impact resistance	1709 Method B	g	3813
Std-OIT		min	23.91
HP-OIT		min	1450

Table 2. Standard properties of LLDPE geomembrane tested.

Properties	Methods (ASTM)	Units	Values
Thickness		mm	0.53
Puncture	D4833	Peak (N)	194.21
Tear resistance	D1004	Peak (N)	53.91 x 55.02
Tensile	D6693	Peak (N)	129.89
Load at break	D6693	(N)	118.28
Elongation at break	D6693	% (min)	955.00
Tensile strength	D882	N	407
Impact resistance	1709 Method B	g	2970
Std-OIT		min	190

The VOCs benzene, toluene, ethylbenzene and xylenes, commonly known as BTEX, were the contaminants examined in this study. These common aromatic hydrocarbons are extremely volatile and often used in VOC migration analysis. They are present in the dissolved and gaseous phase in a range of hazardous settings, including hydrocarbon spills and storage, waste, leachate and landfill gas. Diffusion and sorption tests were performed using BTEX in both a dilute aqueous and vapour phase.

Tests were conducted using purchased BTEX standards and injecting known amounts into vials and cells filled with organic-free distilled deionized water (DD water). In both liquid and vapour sorption tests, liquid samples were taken using gas tight syringes and directly injected into the Purge & Trap syringe filled with distilled deionized water. In the liquid diffusion tests, liquid samples were taken as per the sorption tests. For the vapour diffusion tests, samples were taken from the liquid phase in the cells and directly injected into the P&T syringe. Vapour concentrations were measured from liquid concentration results and then correlating the vapour concentrations in the cells by Henry's Law. Temperatures in the cells were monitored and maintained at 24°C. Surrogates fluorobenzene and 1,4-dichlorobenzene were used as internal standards for quality assurance/quality control. Samples were analyzed by Purge and Trap Gas Chromatography/Mass Spectrometer (P&T)-GC/MS. The procedure used a Hewlett Packard 5890 GC with a P&T unit and 5972 mass selective detector (MS).

3.2 Procedures

3.2.1 Sorption Tests

Single compartment stainless steel cells were secured with a geomembrane sample inside. Liquid sorption tests had cells completely filled with DD water; vapour sorption tests were partially filled with water, leaving an air space. The geomembrane was suspended in the vapour space for these latter tests. BTEX were injected into the liquid phase of all sorption tests. Initial concentrations of the dilute aqueous solution ranged from 0.2-2.0 mg/L (ppm). These concentrations are reflective of typical landfill leachate and landfill gas concentrations (Rowe, 2005; EPA, 2005). Samples were agitated on magnetic stirring plates and maintained at a temperature of 24°C. Contaminant concentrations were evaluated over time until equilibrium was reached. The partitioning coefficient for each contaminant compound was calculated using the following equations derived from the contaminant mass balance at equilibrium:

$$M_{s0} = M_{sF} + M_{gF}, \quad (7)$$

where M_{s0} is the initial mass of the contaminant in solution [M]; M_{sF} is the final mass of the contaminant in solution [M] and M_{gF} is the mass contained in the geomembrane [M]. When Eq. 7 is rearranged in terms of concentrations and volumes, it becomes:

$$c_{f0}V_{f0} = c_{fF}V_{fF} + \frac{M_g}{\rho_g}c_{gF}, \quad (8)$$

where c_{f0} is the initial contaminant concentration in solution [ML^{-3}]; V_{f0} is the initial solution volume [L^3]; c_{fF} is the final contaminant concentration in solution [ML^{-3}]; V_{fF} is the final solution volume [L^3]; M_g is the initial mass of the geomembrane sample [M]; ρ_g is the geomembrane density [ML^{-3}] and c_{gF} is the final concentration of the contaminant in the geomembrane at equilibrium [ML^{-3}]. The partitioning equation can be expressed by substituting Eq. 8 into Eq. 1 to give:

$$S_{gf} = \frac{[c_{f0}V_{f0} - c_{fF}V_{fF}]\rho_g}{M_g c_{gF}} \quad (9)$$

3.2.2 Liquid Diffusion Tests

In the liquid diffusion tests, a stainless steel double compartment cell (source and receptor) was divided by a geomembrane sample. The receptor cell contained DD water. The source cell contained a dilute aqueous BTEX solution ranging between 0.2 and 2.0 mg/L. The liquid phase was sampled to measure the initial concentration in the source. Diffusion cells were maintained at a constant temperature of 24°C. Samples were taken from the source and receptor at regular time intervals until an equilibrium was established.

3.2.3 Vapour Diffusion Tests

In the vapour diffusion tests, a modified stainless steel double compartment cell (source and receptor) was employed. In both the source and receptor compartments, there was an aqueous phase and a vapour phase. A geomembrane sample was secured between the source and the receptor compartments where it rested entirely in the vapour phase. This ensured that the geomembrane sample was solely exposed to contaminants in the vapour phase. The receptor liquid contained DD water. The source liquid contained a dilute aqueous BTEX solution at concentrations ranging from 0.2 to 2.0 mg/L. The chemicals were allowed to equilibrate between the liquid and vapour phase of the source compartment. The liquid phase was sampled to measure the initial concentration in the source and the vapour phase was related using Henry's Law. Diffusion cells were maintained at a constant temperature of 24°C. Samples were taken from the source and receptor liquid phase at regular time intervals.

3.2.4 Modelling Diffusion Parameters

These experiments used theoretical modelling to establish the diffusion (D_g), partition (S_{gf}) and permeation (P_g) coefficients of contaminants through the geomembranes. The concepts and theory of this approach were developed by Rowe et al. (1998) for clayey soils and by Rowe et al. (1995, 1996) for geomembranes. Analysis of experimental results followed the procedure outlined by Rowe et al. (1995) using the finite layer analysis program POLLUTE[®] v.7 (Rowe and

Booker, 2004). This procedure was used in previous studies by Sangam and Rowe (2001) and McWatters and Rowe (2007). Further explanation of the procedures can be found in Sangam and Rowe (2001)

4. RESULTS AND DISCUSSION

4.1 Dissolved Phase Diffusion

Throughout sorption and diffusion tests, contaminant concentrations in the cell (source and receptor) were monitored over time until equilibrium was reached. Theoretical curves obtained with POLLUTEv7 were fitted to the experimental data for both the source and receptor. The sorption coefficient, S_{gf} , was measured from the sorption tests. Both the S_{gf} and diffusion coefficient, D_g , values were obtained from the diffusion tests. Then the permeation coefficient, P_g , for each contaminant was calculated from these curves.

For the diffusion tests in the dissolved phase, Figures 1 and 2, show the decrease in benzene and *m&p*-xylenes concentrations in the source solution, respectively. Figure 3 and 4 show the increase in benzene and *m&p*-xylenes concentrations in the receptor solution. The changes in concentration are plotted as normalized concentrations relative to the initial concentration. The equilibrium concentration was reached after 60 days for benzene and 40 days for *m&p*-xylenes. Benzene concentrations decreased by 58% of the initial concentration, while the decrease in *m&p*-xylenes was 75%. For benzene, the partitioning coefficient was 120 and the diffusion coefficient ranged from $0.6-0.8 \times 10^{-13} \text{ m}^2\text{s}^{-1}$ with the uncertainty that gives rise to the range being a result of the scatter of the experimental data points. *m&p*-Xylenes has an S_{gf} of 430 and a D_g of $0.5 \times 10^{-13} \text{ m}^2\text{s}^{-1}$. Toluene, ethylbenzene and *o*-xylene were also monitored. The results for additional BTEX contaminants were: toluene, $S_{gf}=220$, $D_g=0.6-0.8 \times 10^{-13} \text{ m}^2\text{s}^{-1}$; ethylbenzene, $S_{gf}=425$, $D_g=0.5 \times 10^{-13} \text{ m}^2\text{s}^{-1}$; *o*-xylene, $S_{gf}=400$, $D_g=0.5 \times 10^{-13} \text{ m}^2\text{s}^{-1}$.

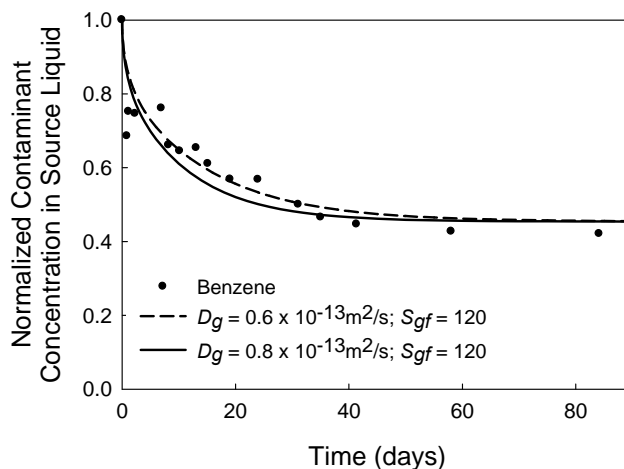


Figure 1. Change in benzene concentration in the source during dissolved phase diffusion tests using co-extruded GM. Experimental data plotted as symbols and theoretical curves plotted as lines.

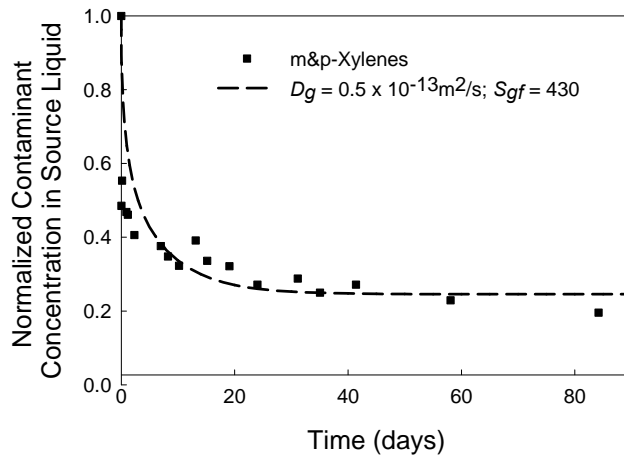


Figure 2. Change in *m*&*p*-xylenes concentration in source during dissolved phase diffusion tests using co-extruded GM.

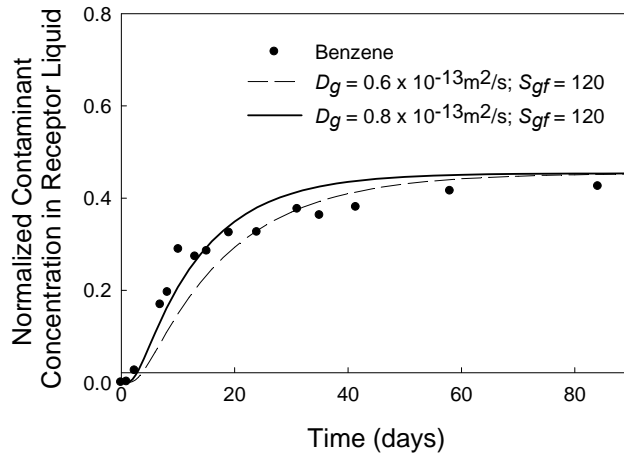


Figure 3. Change in benzene concentration in the receptor during dissolved phase diffusion tests using co-extruded GM.

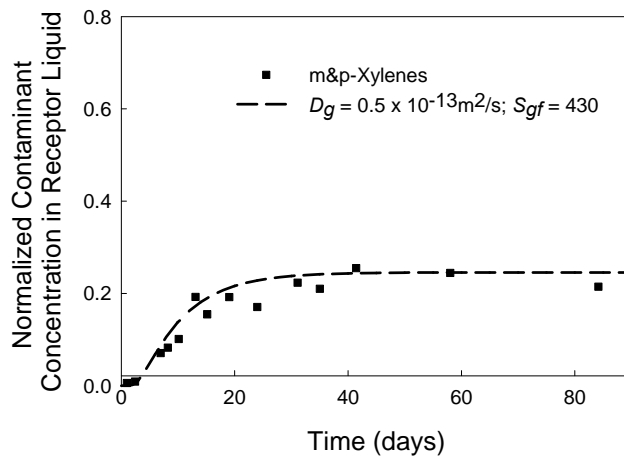


Figure 4. Change in *m*&*p*-xylenes concentration in receptor during dissolved phase diffusion tests using co-extruded GM.

4.2 Vapour Phase Diffusion

For diffusion tests in the gaseous phase, the concentrations of contaminants were measured in the source and receptor liquid phase. Vapour concentrations were calculated using the Henry's Law correlation at 24°C. Temperature was maintained at 24°C and recorded throughout the tests. Figures 5 and 6 show the decrease in benzene and *m&p*-xylenes concentrations in the source vapour throughout the diffusion testing period. The concentrations were normalized relative to the initial concentration of contaminants in the source reservoir. The concentration decrease for benzene in the source was 82% and for *m&p*-xylenes was 91%.

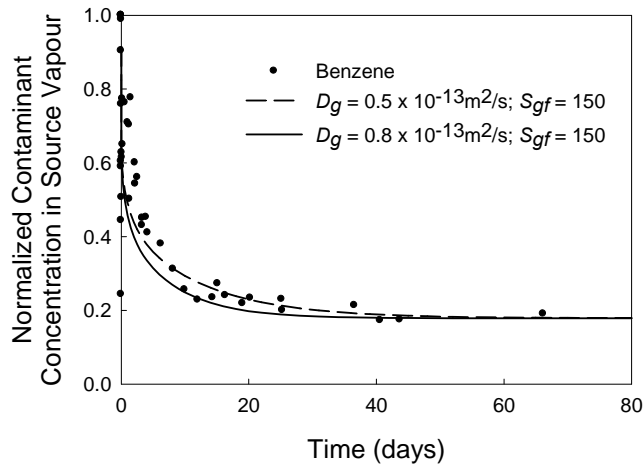


Figure 5. Change in benzene concentration in the source during vapour diffusion tests using co-extruded GM. Experimental data plotted as symbols and theoretical curves plotted as lines.

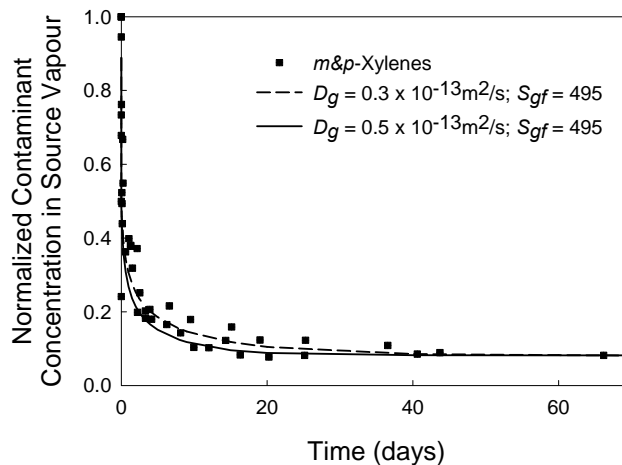


Figure 6. Change in *m&p*-xylenes concentration in the source during vapour diffusion tests using co-extruded GM.

The increase in contaminant concentrations in the receptor are shown in Figure 7 (benzene) and Figure 8 (*m&p*-xylenes). In repeated tests, equilibrium was reached after 55 days for benzene and 40 days for *m&p*-xylenes. For the contaminant benzene, the vapour sorption parameter was 150 and the diffusion coefficient ranged from $0.5\text{-}0.8 \times 10^{-13} \text{ m}^2\text{s}^{-1}$. *m&p*-Xylenes has a sorption parameter of 495 and a diffusion coefficient that ranged from $0.3\text{-}0.5 \times 10^{-13} \text{ m}^2\text{s}^{-1}$.

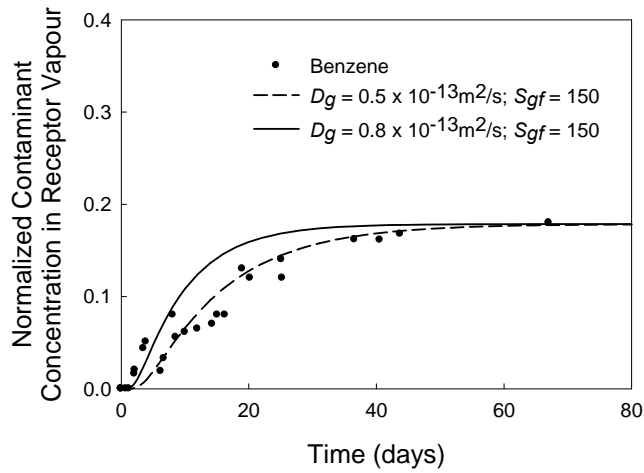


Figure 7. Change in benzene concentration in the receptor during vapour diffusion tests using co-extruded GM.

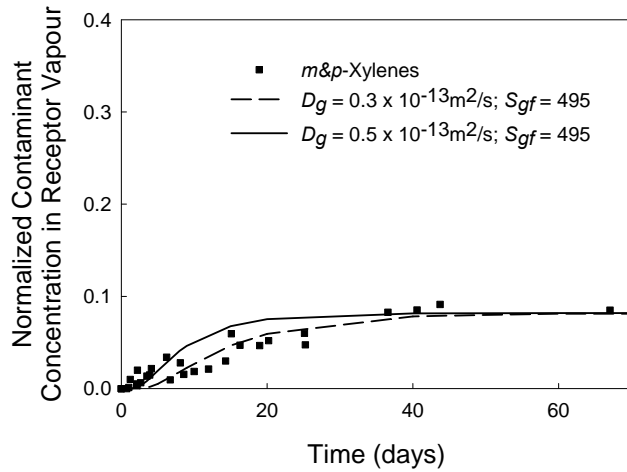


Figure 8. Change in *m&p*-Xylenes concentration in the receptor during vapour diffusion tests using co-extruded GM.

Table 3 presents all three diffusive migration parameters: D_g , S_{gf} and P_g , of all contaminants in both the dissolved and gaseous phase through the co-extruded nylon geomembrane. The partitioning coefficients are slightly higher for the vapour phase than the dissolved phase. The diffusion coefficients for benzene and toluene were similar for both vapour and dissolved phase transport of each contaminant respectively. For the diffusion of ethylbenzene and the xylenes, a diffusion coefficient of $0.5 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ fit with the experimental data. For the vapour phase, a D_g range of $0.3\text{-}0.5 \times 10^{-13} \text{ m}^2 \text{ s}^{-1}$ was necessary to capture the behavior of the gas phase ethylbenzene and xylenes. This is acceptable, considering fluctuations occur between the vapour and liquid phase concentrations as the equilibrium shifts due to slight temperature changes.

Table 3. Inferred partitioning, diffusion and permeation coefficients of the co-extruded nylon GM from diffusion tests in the liquid and vapour phases.

Contaminants	Co-extruded Nylon Liquid Results			Co-extruded Nylon Vapour Results		
	S_{gf}	D_g	P_g	S_{gf}	D_g	P_g
	(-)	($\times 10^{-13} \text{ m}^2\text{s}^{-1}$)	($\times 10^{-10} \text{ m}^2\text{s}^{-1}$)	(-)	($\times 10^{-13} \text{ m}^2\text{s}^{-1}$)	($\times 10^{-10} \text{ m}^2\text{s}^{-1}$)
Benzene	120	0.6-0.8	0.07-0.10	150	0.5-0.8	0.08
Toluene	220	0.5-0.8	0.11-0.18	260	0.4-0.7	0.10-0.13
Ethylbenzene	425	0.5	0.21	485	0.3-0.5	0.15-0.24
<i>m&p</i> -Xylenes	430	0.5	0.22	495	0.3-0.5	0.19-0.25
<i>o</i> -Xylenes	400	0.5	0.20	440	0.3-0.5	0.13-0.22

Sorption and diffusion tests in both the dissolve and vapour states were also performed on the standard 0.53mm (20 mil) LLDPE geomembrane. The results for these parameters (S_{gf} , D_g and P_g) are presented in Table 4, below. The LLDPE geomembrane had a D_g range of $2-4 \times 10^{-13} \text{ m}^2\text{s}^{-1}$ for both the dissolved phase and vapour phases.

Table 4. Inferred partitioning, diffusion and permeation coefficients of standard LLDPE geomembrane from diffusion tests in the dissolved and vapour phases.

Contaminants	LLDPE Liquid Results			LLDPE Vapour Results		
	S_{gf}	D_g	P_g	S_{gf}	D_g	P_g
	(-)	($\times 10^{-13} \text{ m}^2\text{s}^{-1}$)	($\times 10^{-10} \text{ m}^2\text{s}^{-1}$)	(-)	($\times 10^{-13} \text{ m}^2\text{s}^{-1}$)	($\times 10^{-10} \text{ m}^2\text{s}^{-1}$)
Benzene	180	4.0	0.7	150	4.0	0.6
Toluene	350	3.0	1.1	300	2.5	0.8
Ethylbenzene	420	2.0	0.8	420	2.5	1.1
<i>m&p</i> -Xylenes	445	2.0	0.9	440	2.0	0.9
<i>o</i> -Xylenes	400	2.0	0.8	375	2.0	0.8

4.3 Permeation Coefficient Comparisons

As shown in Table 4, the standard LLDPE geomembrane has a permeation coefficient that ranges from $0.7-1.1 \times 10^{-10} \text{ m}^2\text{s}^{-1}$ for BTEXs contaminants in the dissolved phase and a similar range of $0.6-1.1 \times 10^{-10} \text{ m}^2\text{s}^{-1}$ for contaminants in the vapour phase. These permeation coefficients are low, meaning a standard LLDPE geomembrane could provide a reasonable barrier to VOC migration. It would be necessary to perform calculations to establish if the diffusive flux of VOCs through the GM is acceptable for the design purposes of each specific application.

The co-extruded LLDPE with a nylon barrier had a lower permeation coefficient range for both the dissolved phase ($0.07-0.22 \times 10^{-10} \text{ m}^2\text{s}^{-1}$) and vapour phase ($0.08-0.25 \times 10^{-10} \text{ m}^2\text{s}^{-1}$) than the standard LLDPE geomembrane. This is a 5-15 fold decrease in the P_g and S_{gf} values when compared with the standard geomembrane. Therefore, the co-extruded geomembrane would reduce the mass flux of VOC contaminants significantly, providing a better barrier. However, for engineering designs employing geomembranes, consideration should also be given to other factors such as physical properties, long-term performance and cost of each geomembrane.

5. CONCLUSIONS

Diffusion (D_g), partitioning (S_g) and permeation (P_g) coefficients were established for a relatively new co-extruded LLDPE geomembrane with an inner polyamide barrier and a standard LLDPE geomembrane in the dissolved and gaseous phases. P_g values for the 0.38mm (15 mil) co-extruded geomembrane were $0.07-0.22 \times 10^{-10} \text{ m}^2\text{s}^{-1}$ in the dissolved phase and $0.08-0.25 \times 10^{-10} \text{ m}^2\text{s}^{-1}$ in the vapour phase. P_g values for the LLDPE geomembrane were $0.7-1.1 \times 10^{-10} \text{ m}^2\text{s}^{-1}$ (dissolved phase) and $0.6-1.1 \times 10^{-10} \text{ m}^2\text{s}^{-1}$ (vapour phase). The nylon geomembrane showed a 5-15 fold decrease in

both the permeation and diffusion coefficients when compared to the standard LLDPE geomembrane. The co-extruded geomembrane reduced the mass flux of BTEX contaminants. Results from this study show that a co-extruded geomembranes could provide a better barrier to vapour and dissolved phase diffusive migration of VOCs than traditional LLDPE liners of similar thickness when used in containment and barrier systems.

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