



Sorption/desorption, diffusion, permeation and swelling of high density polyethylene geomembrane in the presence of hazardous organic liquids

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Abstract

The laboratory findings on sorption and desorption of 14 organic liquids into high density polyethylene (HDPE) geomembranes are presented at 25, 50 and 70°C. The values of diffusion and permeation coefficients have been calculated using these data. Swelling of the HDPE geomembrane was studied by monitoring its increase in thickness and diameter and thereby calculating the increase in volume. From a temperature dependence of sorption, diffusion and permeation coefficients, the Arrhenius parameters have been calculated. From the sorption data, the concentration profiles of liquids inside the HDPE by solving Fick's diffusion equation under the appropriate initial and boundary conditions. Results of this research may have relevance in the application of HDPE geomembrane in the hazardous waste environment containing the solvents used in this research. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Geomembrane; Sorption; Diffusion; Swelling

1. Introduction

Geomembranes in conjunction with geotextiles or mesh underliners have been used as liners in the containment of hazardous or municipal wastes [1–4]. Even though, many of these are resistant to chemicals, liquids may still permeate at the molecular level and hence their sorption/desorption, diffusion and permeation characteristics are important from the viewpoint of their actual field applications. Recently, National Seal (NSC) in Galesburg, USA has developed several geosynthetic lining systems for solid and liquid

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containment facilities such as hazardous waste ponds, lagoons, reservoirs, etc. Among these, high density polyethylene (HDPE) has been the most widely used as it provides a good chemical resistance and impermeability to many organic liquids [5–12].

Previous papers [13–16], from our laboratory dealt with the solvent resistivity testing of geomembranes. In continuation of this program of research, we now present experimental data on the resistivity of HDPE geomembranes to 14 organic chemicals that are frequently found as leachates in landfill and impoundment sites. Leachate is rather a complex mixture of many different components and it would be difficult to predict their chemical compositions. In order to determine whether a chemical is compatible with a given leachate, one must first identify and chemically characterize the leachate in question. An example of leachate constituent data has been given elsewhere [17]. However, the liquid migration is due to the main organic constituent of the aqueous leachate mixture. Hence, in this study, we have investigated the migration of pure chemicals and their interactions with the HDPE geomembranes. Experimental sorption results have been obtained at 25, 50 and 70°C using a gravimetric method, but the desorption experiments have been carried out at 25°C. From the sorption and desorption results, diffusion coefficients have been calculated using Fick's equation [18]. From the data of sorption, diffusion and permeation coefficients have been calculated. Furthermore, a temperature dependence of sorption, diffusion and permeation has been used to derive the Arrhenius activation parameters. The liquid concentration profiles have also been calculated from a solution of Fick's equation under appropriate initial and boundary conditions using the procedures published earlier [19–21]. The present database might be useful to field engineers while installing the HDPE geomembranes in hazardous chemical pond applications.

2. Theory

The chemical resistance of HDPE geomembrane is related to its ability to prevent the passage of a chemical without destroying it. However, it is possible that the barrier can be breached by chemical interactions which reduce the physical properties of the geomembrane to the point of failure. Even though geomembranes are nonporous materials, liquids, gases and vapors permeate through it on a molecular level and the basic mechanism of transport is essentially the same for all the permeating species. Geomembranes contain interstitial spaces (voids) between the polymer segments through which small molecules can diffuse. Methods have been developed in the literature to estimate the ability of the geomembrane to act as an effective barrier to a chemical [22–24]. Fickian diffusion theory was used to calculate the diffusion coefficient, D , of a liquid into a geomembrane.

According to Fick's law of diffusion, mass transport occurs in the direction of the concentration gradient thereby generating a history of concentration profiles inside the geomembrane. Assuming that the manufacturer provides information on the dangers imposed by certain levels of chemical concentration, it is the task of the engineer to determine the time, duration and value of the concentration profile that may appear in the geomembrane, due to the process of mass transport by molecular diffusion. This is

described by the one-dimensional Fick's equation which can be solved for concentration-independent diffusivity, D , to give [18]:

$$D = \pi \left(\frac{h\theta}{4C_\infty} \right)^2 \quad (3)$$

where C_∞ is the equilibrium concentration at $t \rightarrow \infty$, θ is the slope of the initial linear portion of the sorption/desorption curve and h is thickness of the geomembrane.

Liquid concentration profiles into the geomembrane samples can be calculated using Fick's equation under appropriate initial and boundary conditions to give [19–21]

$$\frac{C_{(x,t)}}{C_\infty} = 1 - \frac{4}{\pi} \sum_{m=0}^{\infty} \frac{1}{(2m+1)} \exp \left[-\frac{D(2m+1)^2 \pi^2 t}{h^2} \right] \sin \left[\frac{(2m+1)\pi x}{h} \right] \quad (4)$$

where m is an integer. By solving Eq. (4), the concentration profiles of the liquids within the geomembrane may be determined. These data are useful to make predictions about the solvent migration inside the geomembrane as a function of time and penetration depth from the face to the middle of the geomembrane.

3. Experimental

3.1. Materials

The hazardous organic liquids considered are: benzene, toluene, *p*-xylene, trimethylbenzene, methoxybenzene, chlorobenzene, 1-chloronaphthalene, dichloromethane, 1,2-dichloroethane, acetone, methyl ethyl ketone, methyl *iso*-butyl ketone, cyclohexanone and butyraldehyde (all were of analytical reagent grade samples supplied from S.D. Fine Chemicals, Mumbai, India, except 1-chloronaphthalene and butyraldehyde which were supplied from Fluka). Some important properties of these liquids are given in Table 1. HDPE was fabricated at the NSC Research Center in Galesburg, USA (courtesy of Mr. J. Donaldson and Mr. J. Siebken) in sheets of dimensions, 35 cm \times 30 cm \times 0.16 cm. Some typical properties of HDPE are summarized in Table 2.

3.2. Immersion tests

In an immersion experiment, the geomembrane is exposed to the chemical for a definite time and the change in mass or dimension of the samples is measured. In tests in which the mass is accurately monitored as a function of time, diffusion and sorption coefficients of the chemical in the geomembrane have been calculated using the procedures published earlier [22–24]. Sorption experiments were performed at 25, 50 and 70°C in an air circulating electronic oven (WTB Binder, Germany) within the accuracy of $\pm 0.5^\circ\text{C}$. The circularly cut disc-shaped geomembrane samples (diameter \approx 2.00 cm) were conditioned in a vacuum oven at 25°C for at least 48 h before experimentation. These samples were then exposed to about 15–20 ml of liquids kept

Table 1
Some physical properties of liquids: molar volume (V), dipole moment (μ), solubility parameter (δ)

Liquids	Chemical formula	V_s (cm ³ /mol)	$\mu(D)$	δ (J cm ⁻³) ^{1/2}
Benzene	C ₆ H ₆	89.4	0.00	9.17
Toluene	C ₆ H ₅ CH ₃	106.9	0.31	8.91
<i>p</i> -Xylene	C ₆ H ₄ (CH ₃) ₂	123.9	0.02	8.77
Trimethylbenzene	C ₆ H ₃ (CH ₃) ₃	139.6	0.00	8.80
Methoxybenzene	C ₆ H ₅ OCH ₃	109.3	1.25	9.50
Chlorobenzene	C ₆ H ₅ Cl	102.2	1.62	9.68
1-Chloronaphthalene	C ₁₀ H ₇ Cl	137.7	1.33	a
Dichloromethane	CH ₂ Cl ₂	64.5	1.14	9.88
1,2-Dichloroethane	C ₂ H ₄ Cl ₂	79.4	1.83	9.78
Acetone	CH ₃ COCH ₃	74.0	2.69	10.00
Methyl ethyl ketone	CH ₃ CH ₂ COCH ₃	90.2	2.76	9.30
Methyl <i>iso</i> -butyl ketone	(CH ₃) ₂ CHCH ₂ COCH ₃	125.8	a	a
Cyclohexanone	C ₆ H ₁₀ CO	104.7	3.80	9.90
Butyraldehyde	CH ₃ (CH ₂) ₂ CHO	89.6	a	0.43

a—data not available.

inside the screw-tight test bottles maintained at the desired temperature ($\pm 0.5^\circ\text{C}$). The mass measurements were done at suitably selected time intervals by removing the samples and then wiping the surface adhering liquid drops using filter-papers. These were then placed on a top-loading digital Mettler balance (Model AE 240, Switzerland, sensitive to ± 0.01 mg) to measure the mass uptake, C_t using the following equation.

$$C_t = \left(\frac{W_t - W_0}{W_0} \right) \frac{1}{M} \times 100 \quad (1)$$

where W_0 is initial mass of the sample, W_t is its mass at time t , for the immersion period and M is the molar mass of the solvent under consideration.

Table 2
Some typical properties of HDPE geomembrane

Property	Method	
Thickness	ASTM D 751, NSF mod.	1.59 mm
Density	ASTM D 1505	0.948 g/cm ³
Carbon black content	ASTM D 1603	2.35%
Tensile properties	ASTM D 638	
Stress at yield		17.6 MPa
Stress at break		33.4 MPa
Strain at yield	1.3" gauge length (NSF)	16.9%
Strain at break	2.0" gauge or extensometer	890%
Modulus of elasticity	ASTM D 638	931 MPa
Dimensional stability	ASTM D 1204, NSF mod.	0.4%
Tear Resistance	ASTM D 1004	1050 N/cm
Puncture Resistance	ASTM D 4833	3728 N/cm
Water Absorption	ASTM D 570 at 23°C	0.05%
Water Vapor Transmission	ASTM E 96	0.009 g/day m ²

Desorption measurements were performed by keeping the already soaked samples in a vacuum controlled oven previously set at 25°C. The mass loss of the samples was monitored at regular intervals of time by removing them from the oven and weighing in the same manner as was done in sorption experiments. When the samples attained equilibrium desorption, no more mass loss occurred and this did not change significantly over a further period of 1 or 2 days. The mol% decrease in concentration after desorption was then calculated as:

$$C_t = \left(\frac{W_t^d - W_0^d}{W_0^d} \right) \frac{1}{M} \times 100 \quad (2)$$

where W_t^d refers to mass of the desorbed sample at time, t and W_0^d is mass of the polymer after complete desorption, i.e. the original mass of the sample.

4. Results and discussion

4.1. Sorption / desorption kinetics

The mol% sorption results, i.e., plots of C_t vs. $t^{1/2}$ curves for the aromatic liquids at 25, 50 and 70°C with the HDPE geomembrane are presented in Fig. 1. In the case of benzene and other substituted benzenes at 25°C, the sorption curves show sigmoidal behavior suggesting the slight deviations from Fickian mechanism. However, such sigmoidal shapes are not so significant at 50 and 70°C. For methoxybenzene, 1-chloronaphthalene and trimethylbenzene, the sorption curves are lower than the other aromatic liquids considered. From the desorption curves at 25°C included in Fig. 1, it is observed that benzene desorbs much faster than 1-chloronaphthalene. The sorption results of aliphatic liquids at 25, 50 and 70°C are presented in Fig. 2. At 25°C, except for dichloromethane and 1,2-dichloroethane, the remaining liquids exhibit lower values of equilibrium sorption ranging between 0.01 and 0.03 mol%. At 50°C, the trends are quite different than those observed at 25°C. For example at 25 and 50°C, butyraldehyde exhibits a slight decrease in sorption after reaching the equilibrium. At 70°C, the equilibrium curves for methyl *iso*-butyl ketone and methyl ethyl ketone vary almost identically and hence their dependencies are shown by a single common curve. At 70°C, 1,2-dichloroethane exhibits a step function after attaining the equilibrium. The desorption curves at 25°C for the aliphatic liquids are also included in Fig. 2; wherein, desorption is quite fast for dichloromethane, but very slow for cyclohexanone. Observation of the mol% uptake data presented in Table 3 suggests that HDPE geomembrane is more resistive toward aliphatic liquids than the aromatic liquids.

The initial sorption results (before completion of 50–55% of equilibrium) have been analyzed using the relationship [25,26]:

$$\frac{C_t}{C_\infty} = Kt^n \quad (5)$$

The values of K represent the extent of interactions between the liquids and the HDPE geomembrane. The parameter values of K and n have been obtained by the method of

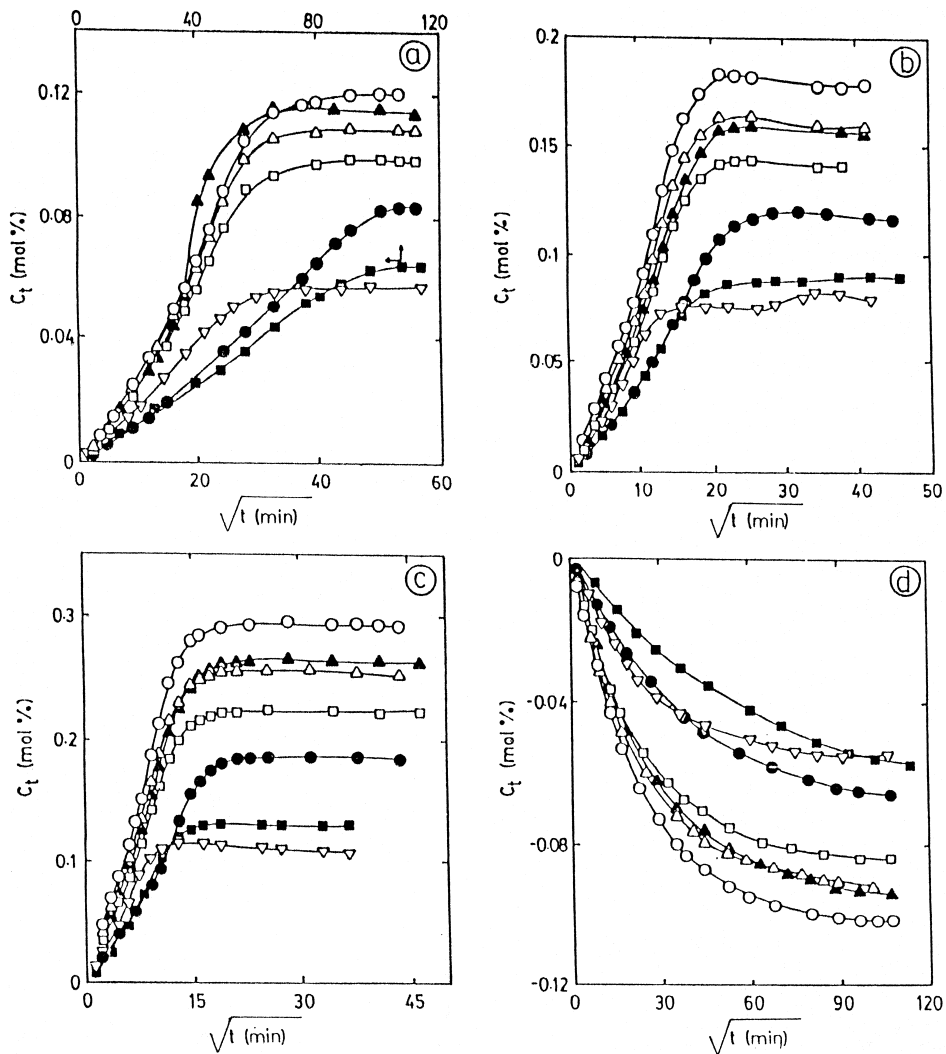


Fig. 1. Sorption curves, i.e., mol% uptake (C_t) vs. square root of time ($t^{1/2}$) for HDPE geomembrane with (○) benzene, (△) toluene, (□) *p*-xylene, (●) trimethylbenzene, (▲) chlorobenzene, (■) 1-chloronaphthalene and (▽) methoxybenzene at (a) 25°C, (b) 50°C, (c) 70°C, and (d) desorption curves at 25°C.

least-squares, but only the results of K are presented in Table 4. The values of K increased systematically with increasing temperature. The values of n vary from 0.50 to 0.61 indicating an anomalous transport [22–24].

4.2. Diffusion and permeation

A number of investigators have studied the solvent resistivity of HDPE geomembranes [5–12]. Values of D from the literature are compared with the present data in

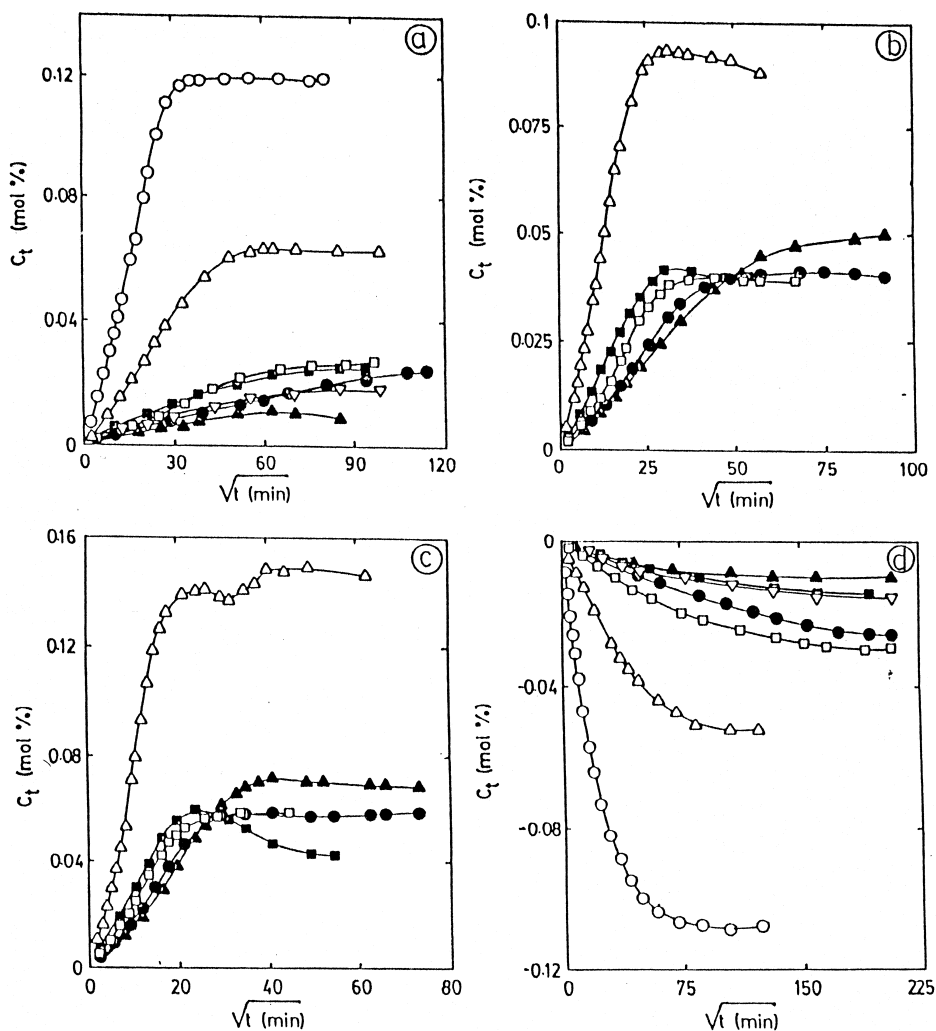


Fig. 2. Sorption curves, i.e., mol% uptake (C_t) vs. square root of time ($t^{1/2}$) for HDPE geomembrane with (○) dichloromethane, (Δ) dichloroethane, (□) methylethyl ketone, (●) methyl *iso*-butyl ketone, (▲) cyclohexanone, (■) butyraldehyde and (▽) acetone at (a) 25°C, (b) 50°C, (c) 70°C, and (d) desorption curves at 25°C.

Table 5. These values provide a starting point, but they should be used with caution because these depend on temperature and concentration. Additionally, the values may also vary depending upon the chemical composition of the containment, polymer crystallinity, additives, etc. and hence, published data should only be considered as an initial guide, but may not replace the experimentally determined values where uncertainty regarding diffusion coefficient may have a significant impact.

Permeability coefficients, P have been calculated from the relation: $P = DS$ and the data are included in Table 4. It is found that the values of P follow the same trends as

Table 3

Sorption (S in mol%) and diffusion coefficients (D 10^7 cm²/s) for HDPE geomembrane with organic liquids

Liquids	S (mol%)			D (10^7 cm ² /s)		
	25°C	50°C	70°C	25°C	50°C	70°C
Benzene	0.12	0.18	0.30	0.64	1.84	3.22
Toluene	0.11	0.17	0.26	0.68	2.40	3.62
<i>p</i> -Xylene	0.10	0.15	0.22	0.64	2.28	3.60
Trimethylbenzene	0.08	0.12	0.19	0.34	1.17	1.81
Methoxybenzene	0.06	0.08	0.12	0.21	0.94	1.68
Chlorobenzene	0.12	0.16	0.27	0.54	1.59	3.07
1-Chloronaphthalene	0.06	0.09	0.13	0.07	0.43	0.90
Dichloromethane	0.12	a	a	0.82	a	a
1,2-Dichloroethane	0.06	0.09	0.15	0.42	1.49	2.35
Acetone	0.02	a	a	0.17	a	a
Methyl ethyl ketone	0.03	0.04	0.06	0.19	0.73	1.50
Methyl <i>iso</i> -butyl ketone	0.03	0.04	0.06	0.07	0.37	1.03
Cyclohexanone	0.01	0.05	0.07	0.16	0.29	0.60
Butyraldehyde	0.02	0.04	0.06	0.34	1.14	1.72

those of D . However, the permeation coefficients of the present liquids are quite lower further suggesting the high resistivity of HDPE geomembrane to these liquids. The values of D and the K obtained from desorption experiments at 25°C are presented in Table 6. Except in a few cases, the D and K values are lower than those observed for sorption. This might be attributed to the different drying mechanisms included especially by the higher boiling liquids.

Table 4

Results of parameter K (in g/g(mol) ^{n}) of Eq. (5) and permeation coefficients (P 10^7 cm²/s) for HDPE geomembrane with organic liquids

Liquids	K (in g/g(mol) ^{n})			P (10^7 cm ² /s)		
	25°C	50°C	70°C	25°C	50°C	70°C
Benzene	1.68	4.37	7.26	0.06	0.26	0.74
Toluene	1.72	4.19	7.01	0.07	0.37	0.82
<i>p</i> -Xylene	1.91	4.01	6.57	0.07	0.36	0.86
Trimethylbenzene	1.24	2.56	4.62	0.03	0.17	0.41
Methoxybenzene	1.93	3.12	4.76	0.01	0.08	0.21
Chlorobenzene	1.75	3.86	5.67	0.07	0.29	0.92
1-Chloronaphthalene	1.41	1.88	2.66	0.01	0.06	0.19
Dichloromethane	2.40	a	a	0.08	a	a
1,2-Dichloroethane	1.22	2.77	3.27	0.03	0.14	0.35
Acetone	1.90	a	a	0.002	a	a
Methyl ethyl ketone	0.68	1.74	3.41	0.004	0.02	0.06
Methyl <i>iso</i> -butyl ketone	1.43	1.13	2.50	0.002	0.02	0.06
Cyclohexanone	2.88	1.51	1.85	0.002	0.01	0.04
Butyraldehyde	2.95	3.01	5.25	0.006	0.04	0.08

Table 5

Literature comparison of diffusion coefficients for containments in HDPE geomembrane at 25°C

Containment	Thickness (mm)	D (10^{11} cm ² /s)		References
		Literature	Present	
Benzene	1.50	0.22–0.33	0.64	[11]
Toluene	1.50	0.26–0.40	0.68	[11]
Chlorobenzene	1.50	0.22–0.32	0.54	[11]
Dichloromethane	2.00	0.2–1.0	0.82	[29]
1,2-Dichloroethane	2.00	0.3–0.6	0.42	[8]
Acetone	2.00	0.026–0.1	0.17	[30]
Methyl ethyl ketone	2.50	0.075	0.19	[30]

The calculated concentration profiles from Eq. (4) for the high diffusing benzene and the low diffusing 1-chloronaphthalene at 50°C are displayed in Fig. 3. In all cases, while generating the theoretical curves, we could not use the same initial times, but when we compared the profile values for different liquids, the high-diffusing benzene attains equilibrium sorption much faster than the low-diffusing 1-chloronaphthalene.

4.3. Temperature effects—Arrhenius activation parameters

Sorption, diffusion and permeation coefficients show an increase with increasing temperature. This prompted us to calculate Arrhenius parameters for the transport processes. Our present data showed linear relationships in the plots of $\ln D$, $\ln S$ or $\ln P$ vs. $1/T$ suggesting that the Arrhenius relationship is applicable to derive the activation parameters E_D , E_P and ΔH_S for the process of diffusion, permeation and sorption

Table 6

Diffusion coefficients (D) and the parameter (K) for the desorption of organic liquids with HDPE geomembrane

Liquids	D (10^7 cm ² /s)	K (10^2 g/g(min) ⁿ)
Benzene	0.76	3.69
Toluene	0.70	3.18
<i>p</i> -Xylene	0.68	3.10
Trimethylbenzene	0.30	1.76
Methoxybenzene	0.16	2.16
Chlorobenzene	0.61	2.25
1-Chloronaphthalene	0.08	0.47
Dichloromethane	0.17	9.17
1,2-Dichloroethane	0.23	2.09
Acetone	0.09	3.18
Methyl ethyl ketone	0.11	0.51
Methyl <i>iso</i> -butyl ketone	0.05	0.70
Cyclohexanone	0.16	1.14
Butyraldehyde	0.07	1.79

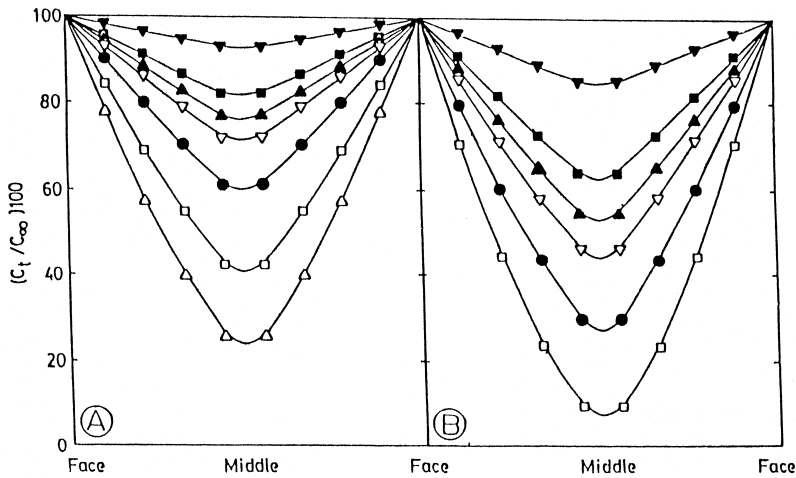


Fig. 3. Concentration profiles calculated from Eq. (4) for (A) benzene and (B) 1-chloronaphthalene for (Δ) 4.00 min, (\square) 8.00 min, (\bullet) 20.00 min, (∇) 40.00 min, (\blacktriangle) 60.00 min, (\blacksquare) 100.00 min, (\blacktriangledown) 600.00 min with HDPE geomembrane at 50°C.

respectively. These parameters have been obtained by fitting the data to linear Arrhenius equation of the type:

$$X = X_0 e^{-E_x/RT} \quad (6)$$

where $X = S, D,$ or P ; $X_0 = S_0, D_0$ or P_0 ; RT has the usual meaning; the parameters E_D, E_P and ΔH_S thus obtained by the least-squares procedure are presented in Table 7. The values of E_D and E_P are generally higher for all the liquids suggesting the extra

Table 7

Activation energy for diffusion (E_D in kJ/mol), permeation (E_P in kJ/mol), heat of sorption (ΔH_S in kJ/mol), interaction parameter, χ and swelling index, α (cm^3/g) for HDPE geomembrane with organic liquids

Liquid	E_D	E_P	ΔH_S	χ	α
Benzene	30.9 ± 2.3	47.6 ± 0.2	16.7 ± 2.4	0.36	0.10
Toluene	32.3 ± 5.9	48.5 ± 4.4	16.2 ± 1.5	0.34	0.11
<i>p</i> -Xylene	33.1 ± 5.4	48.4 ± 3.5	15.3 ± 1.9	0.34	0.12
Trimethylbenzene	32.1 ± 5.3	48.8 ± 3.7	15.3 ± 2.2	0.34	0.11
Methoxybenzene	40.2 ± 6.1	53.4 ± 4.0	13.3 ± 1.1	0.44	0.06
Chlorobenzene	33.1 ± 1.3	48.5 ± 2.4	15.4 ± 3.7	0.48	0.11
1-Chloronaphthalene	48.0 ± 6.3	61.3 ± 4.4	13.4 ± 0.9	a	0.08
Dichloromethane	a	a	a	0.47	0.03
1,2-Dichloroethane	33.0 ± 5.3	49.2 ± 2.8	16.7 ± 2.3	0.48	0.05
Acetone	a	a	a	0.53	0.01
Methyl ethyl ketone	39.6 ± 2.9	52.7 ± 0.4	13.2 ± 1.9	0.38	0.02
Methyl <i>iso</i> -butyl ketone	51.0 ± 1.7	65.7 ± 2.0	14.7 ± 0.3	a	0.01
Cyclohexanone	24.9 ± 3.9	57.2 ± 0.9	32.4 ± 4.9	0.57	0.01
Butyraldehyde	31.3 ± 5.6	49.7 ± 6.1	19.5 ± 1.1	0.34	0.02

a—data not obtained.

energy needed to cross-over the Eyring-type potential energy barrier indicating that HDPE is a rigid structure and is highly resistive for the liquids used. The ΔH_s values are generally small and vary around 15 kJ/mol, except cyclohexanone for which $\Delta H_s = 32$ kJ/mol. The positive values of ΔH_s suggest that the sorption follows an endothermic process. Furthermore, the positive values of ΔH_s suggest that the heat of condensation is positive and greater in magnitude than the heat of mixing $\Delta H_{\text{mix}} = \Delta H_s + \Delta H_v$, where H_{cond} depends on heat of vaporization of the liquid, ΔH_v so that: $\Delta H_s = H_{\text{cond}} + \Delta H_{\text{mix}}$. However, ΔH_{mix} is a function of polymer–solvent interaction parameter, χ which can be calculated as [27]:

$$\chi = \beta + \frac{V_s}{RT} (\delta_s - \delta_p)^2 \quad (7)$$

Here, δ_s and δ_p are, respectively, the solubility parameters of solvent and polymer, β is a lattice constant, the value of which is generally taken as 0.34, and the term RT has the usual meaning.

In order to calculate χ , a prior knowledge of the solubility parameter of the polymer, δ_p is needed. The procedure suggested by Gee [28] was used to compute δ_p . A plot of swelling parameter $\alpha = [(M_a - M_b)/M_b \rho_s]$ vs. δ_s (where M_a and M_b are, respectively, the mass of geomembrane after and before solvent sorption; ρ_s is density of the solvent) was constructed and δ_p was obtained from the maximum value of α which was observed at a δ_s value of $9.17 \text{ (J/cm}^3\text{)}^{1/2}$ for the HDPE geomembrane. Using these values in Eq. (7), the results of χ have been calculated and these values are included in Table 7 along with the results of α . It is found that the values of χ range from 0.34 to 0.57, an expected range of the rubbery polymer–solvent systems. These values are generally lower for aromatic liquids than aliphatics. Similarly, the α values are lower for aliphatics than aromatics.

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