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Sorption and Diffusion of Organic Liquids into Fluoroelastomer Membranes

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ABSTRACT

Sorption and diffusion of organic liquids into fluoropolymer (FC-2179) membranes have been investigated from 30 to 60°C using a gravimetric method. Diffusion coefficients, percent mass uptake, and apparent activation energies for the transport processes have been estimated. Diffusion coefficients of the liquids into the membrane have been computed from Fick's relation. A Flory–Huggins-type interaction parameter was obtained from the solubility parameter concept. Furthermore, the activation parameter values and heat of sorption data have been studied in terms of heat of mixing. The values of diffusion coefficients did not show any considerable dependence on solvent concentration. However, solvent transport as analyzed from an empirical equation was found to be of the anomalous type. Molecular transport also showed a dependence on the chemical nature of the liquids. The concentration profiles of liquids have been calculated at different penetration depths of the membrane at different time intervals by solving Fick's differential equation under suitable initial and boundary conditions. A numerical method based on the finite difference technique was also used to predict the concentration profiles of liquids, and these are compared with the profiles computed from an analytical solution of Fick's equation.

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INTRODUCTION

Membrane-based separation processes (MBSPs) have potential applications in the effective recycling of hazardous wastes generated in chemical processing industries, which otherwise would pose serious environmental pollution problems (1–3). Among many available MBSPs, the primary application of reverse osmosis (RO) has been the desalination of brackish water and seawater (4). Due to improvements in RO technology, a host of other applications have been studied (5–7). Other MBSPs are being used in such diverse areas as metal finishing and biotechnology (8). In all MBSPs, a key factor to understanding molecular transport phenomenon and application of membranes involves the assessment of sorption and diffusion parameters. With regard to organic–organic or organic–aqueous liquid mixture separation, the pervaporation (PV) separation technique has played a major role in many chemical industries. A recent review by Aminabhavi et al. (9) addresses several aspects of the PV technique.

High performance fluoroelastomers developed by 3M Chemical Products Division, USA, compete against thermosets as versatile matrix materials in several engineering applications (10). Thermoplastics offer advantages such as high fracture toughness and ease of processing when compared to thermosets. However, in applications requiring exposure to aggressive liquids and vapors, their chemical resistance is important. In the present paper, the Fluorel 3M fluoroelastomer (FC-2179) membrane is chosen as the candidate material and its interactions with aromatic liquids are studied. FC-2179 is a copolymer of tetrafluoroethylene and propylene with 65% fluorine. It is known to exhibit excellent chemical resistivity and mechanical strength even under severe application conditions. The solvent transport characteristics, such as sorption and diffusion of organic liquids, have not been studied previously.

Solvents used in this study as penetrants are: 1,2-dichloroethane, 1,1,1-trichloroethane, benzene, toluene, methoxybenzene, and nitrobenzene. Sorption experiments were performed at 30, 45, and 60°C using a gravimetric method. Diffusion coefficients were calculated from Fick's equation (11). Furthermore, the concentration profiles of liquids were calculated by solving Fick's diffusion equation and also by using a numerical method based on the finite difference technique (12). From the temperature dependence of transport coefficients, the apparent values of Arrhenius activation parameters were calculated. Experimental and calculated results are discussed in terms of polymer–solvent interactions. The present paper is a continuation of our ongoing program of research activity on the study of small molecule transport into engineering polymer membranes (13–16).

EXPERIMENTAL

Research grade samples of 1,2-dichloroethane, 1,1,1-trichloroethane, benzene, and toluene were purchased from s.d. fine Chem. Pvt. Ltd. (Mumbai, India); methoxybenzene and nitrobenzene were obtained from Thomas Baker Pvt. Ltd. (Mumbai, India). The solvents were procured in their highest purity and were used without further purification. Some important properties of the solvents are listed in Table 1.

Fluorel FC-2179 was milled or internally mixed with standard fillers and other ingredients during formulations. The "dry" ingredients were blended before being added to the masticated gum. Once mixed, the compounded stocks display excellent processing safety and storage stability. Press cure conditions of 175 to 205°C for 3 to 5 minutes are recommended with a postcure cycle of 16 to 24 hours at 230 to 260°C in order to produce the optimum physical properties. Representative properties of the FC-2179 membranes are listed in Table 2.

FC-2179 grade fluoroelastomer sample slabs with dimensions of 55 cm × 30 cm and initial thicknesses ranging from 0.219 to 0.231 cm were used. From these, circular disk-shaped samples with a diameter of 1.980 cm were cut using a sharp-edged carbon-tipped steel die. The cut samples were dried in vacuum desiccators over anhydrous calcium chloride at room temperature at least 24 hours before the start of the sorption experiments. The samples were first soaked in screw-tight test bottles containing 15–20 mL of the solvents maintained at the desired temperature (±0.5°C) in an electronically controlled hot air incubator (WTB Binder, Germany). Polymer samples were removed periodically; the solvent drops adhering

TABLE 1
Physical Properties of Organic Liquids Used as Penetrants

Liquid	Chemical formula	Molar volume ($cm^3 \cdot mol^{-1}$)	Dipole moment (μ)	Solubility parameter (δ_S) $(J \cdot cm^{-3})^{1/2}$
1,2-Dichloroethane	$CH_2Cl—CH_2Cl$	79.4	1.83	9.95
1,1,1-Trichloroethane	CH_3CCl_3	100.0	1.70	9.69
Benzene	C_6H_6	89.4	0.00	18.74
Toluene	$C_6H_5CH_3$	106.9	0.31	18.23
Methoxybenzene	$C_6H_5OCH_3$	109.3	1.83	19.50
Nitrobenzene	$C_6H_5NO_2$	102.7	4.00	20.50

TABLE 2
Compound Formulations and Some Representative Physical/Mechanical Properties of Fluoroelastomer

Compounds	Wt%
MT Black (N-990)	30
MgO (Magnalite D)	3
$Ca(OH)_2$	6
Typical Rheological Properties	
Mooney scorch, MS at 121°C	
Initial reading	100
Minimum reading	75
Minutes to 10 point rise	25+
Monsanto rheometer, 177°C, 100 counts/min, 3° arc	
Minimum torque, Nm	3.1
Time to 1 Nm rise, minutes	4.1
Time to 50 Nm torque, minutes	6.5
Maximum torque at 12 minutes, Nm	13.9
Typical Mechanical Properties	
Press cure, 5 minutes at 177°C	
Tensile strength, Nm^{-2}	238.6
Elongation, %	200
Hardness, Shore A	75
100% Modulus, Nm^{-2}	108
Aged 70 hours at 276°C (air)	
Tensile strength, Nm^{-2}	186.4
Elongation, %	195
Hardness, Shore A	67
Volume change, %	+18
Compression set, %, Method B (0.139 inch O-ring, ASTM D-395)	
70 hours at 24°C	8
70 hours at 200°C	12
336 hours at 200°C	28

to the surface were wiped off using the filter paper wraps and weighed immediately on a digital Mettler balance (Model AE 240, Switzerland) within a precision of ± 0.01 mg. Other experimental details were the same as those given earlier (13–16).

RESULTS AND DISCUSSION

Fick's second law for one-dimensional diffusion relates the changes in concentration as a function of time to the change in solvent flux with

respect to position (11):

$$\frac{\partial C}{\partial t} = D\left(\frac{\partial^2 C}{\partial x^2}\right) \tag{1}$$

where C is the concentration, t is the time, x is a position point along the axis, and D is the diffusion coefficient. The solution of Eq. (1) for a plane sheet of thickness h is given by

$$\frac{M_t}{M_\infty} = \left(\frac{4D^{1/2}}{\pi^{1/2}h}\right) t^{1/2} \tag{2}$$

where M_t is the polymer weight uptake at time t, and M_∞ is the equilibrium weight uptake. The concentration-independent diffusion coefficients D of the polymer–solvent systems were calculated from the slopes of the initial linear portions of the sorption curves. The values thus calculated are accurate to ± 0.01 units of the diffusion coefficients. In the present investigation the results of D are considered to be constant as evidenced by the fact that the sorption curves are nonsigmoidal, thus suggesting Fickian transport.

The values of D presented in Table 3 show a dependence on temperature as well as on the nature of the liquids. The diffusion coefficients of the two haloalkanes serve as examples of molecules exhibiting the limiting behaviors of all the liquids used. For instance, 1,2-dichloroethane has the highest values of D at all temperatures, whereas 1,1,1-trichloroethane shows the lowest D compared to the other liquids. However, intermediate values are shown by the aromatic liquids. Among the latter class of molecules, nitrobenzene at 45 and 60°C shows higher values than the other aromatics, but at 30°C it exhibits the smallest D value of 0.71×10^{-8} $cm^2 \cdot s^{-1}$. Toluene, though it has a higher molar volume than benzene, exhibits higher values of D except at 30°C. The diffusion coefficients of toluene and methoxybenzene at 30°C are identical, but at higher temperatures the diffusion coefficients of toluene are higher than those of methoxybenzene. It may be noted that the diffusion coefficients of the present systems do not seem to be dependent in any systematic manner with such physical characteristics as the dipole moment or the solubility parameter values of the liquids.

Figure 1 shows the weight uptake values of liquids as a function of the square root of time at 30 and 60°C. It is observed that the sorption is affected by temperature as well as by the nature of the liquids used as penetrants. For instance, at 30°C the weight uptake curves initially show a linear dependence on the square root of time, and the time to attain equilibrium sorption is smaller at 30°C than at 60°C. In all cases the equilib-

TABLE 3
Equilibrium Sorption (S), Diffusion Coefficients (D), and Estimated Parameter K of Eq. (3) for Fluoroelastomer Membrane with Organic Liquids

Liquid	S (wt%)			$D \times 10^8$ ($cm^2 \cdot s^{-1}$)			$K \times 10^2$ ($g/g \cdot min^n$)		
	30°C	45°C	60°C	30°C	45°C	60°C	30°C	45°C	60°C
1,2-Dichloroethane	13.53	15.69	18.74	4.38	9.95	21.38	1.68	2.34	2.69
1,1,1-Trichloroethane	12.74	15.81	18.90	0.62	2.43	6.66	0.73	1.00	1.43
Benzene	8.72	11.86	13.32	3.73	5.68	8.57	1.27	1.90	1.83
Toluene	9.80	10.43	11.873	2.04	6.20	12.68	1.06	1.60	2.39
Methoxybenzene	14.51	14.88	15.88	2.04	5.73	7.36	1.76	1.73	1.75
Nitrobenzene	6.89	34.06	37.17	0.71	6.70	13.66	1.26	1.64	2.14

rium sorption increases with temperature, suggesting an increased molecular migration of liquids due to the possible creation of extra free volume spaces within the polymer matrices. At 30°C, methoxybenzene shows a higher S than all the other liquids. On the other hand, nitrobenzene at 30°C shows smaller values of S when compared to the other liquids; however, at 60°C its sorption values have increased considerably and these values supersede all the other liquids. Between benzene and toluene, the latter exhibits a lower S at 30°C than the former. At 60°C the sorption values of benzene are higher than those for toluene. The sorption behaviors of 1,2-dichloroethane and 1,1,1-trichloroethane are almost identical.

In order to test whether or not sorption follows the Fickian mode, the dynamic sorption results before completion of 55% of equilibrium were fitted to the empirical relationship (11)

$$M_t/M_\infty = Kt^n \tag{3}$$

where K is a parameter characteristic of the polymer–solvent system. The value of n indicates the type of transport. For Fickian transport, $n = 0.5$, while $n = 1.0$ for Case II or non-Fickian transport. Values of n between 0.5 to 1.0 define anomalous transport. The values of n and K have been calculated from the least-squares method by fitting the sorption results to Eq. (3). It is found that n ranges between 0.50 and 0.54 in all cases, suggesting anomalous transport. The values of K are also included in Table 3. The values for 1,1,1-trichloroethane are lower, but the K values of 1,2-dichloroethane are higher than for other liquids, suggesting its higher interaction with polymer chain segments. The results of K increase systematically with increasing temperature in all the polymer–solvent systems.

The sorption results presented in Table 3 show a clear-cut dependence on the physical properties and structural characteristics of the liquids.

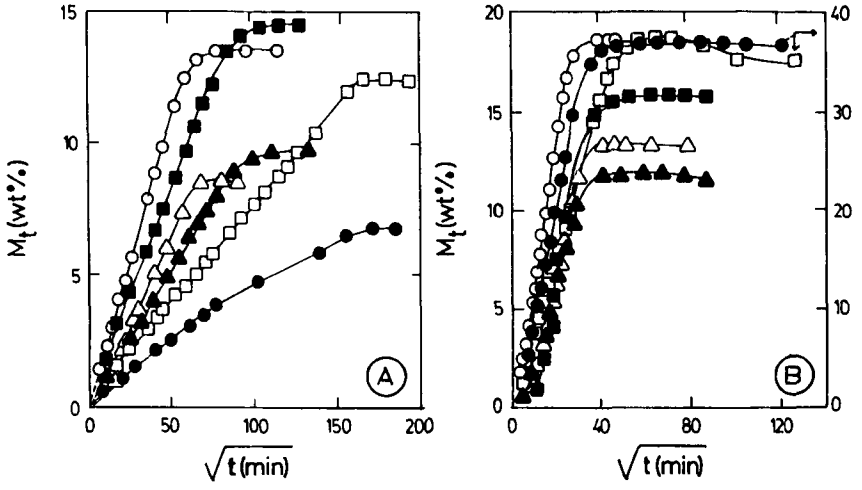

FIG. 1 Sorption curves of the polymer with organic liquids at (A) 30°C and (B) 60°C. (○) 1,2-Dichloroethane; (□) 1,1,1-trichloroethane; (△) benzene; (▲) toluene; (■) methoxybenzene; and (●) nitrobenzene.

According to Hildebrand (17), the solubility parameter concept can be used to explain the sorption results, but the present S values do not show any significant dependence on differences in the solubility parameter, i.e., δ_S values of the liquids. For instance, benzene and toluene, which have almost identical values of δ_S, also exhibit similar sorption tendencies. Similarly, 1,2-dichloroethane and 1,1,1-trichloroethane exhibit almost identical sorption values at all temperatures; it may be noted that the δ_S and dipole moment (μ) values of these liquids are not too different (see Table 1). However, nitrobenzene, with a δ_S values of 20.5 $(J \cdot cm^{-3})^{1/2}$, exhibits a lower S at 30°C but higher S values at 45 and 60°C. On the other hand, methoxybenzene, which has a δ_S value of 19.5 $(J \cdot cm^{-3})^{1/2}$, shows negligible variations in S with differences in temperature. Thus, for the aromatic liquids, molecular transport seems to depend on the nature of the substituents on the benzene moiety. For instance, nitrobenzene with a μ value of 4.0 exhibits a lower S at 30°C, but at 45 and 60°C its sorption values are exceedingly higher than all the other substituted benzenes. Similarly, toluene, which is a slightly more polar molecule than benzene, shows higher sorption at 30°C but not at higher temperatures.

In order to explain the sorption results, further attempts were made to calculate the Flory–Huggins-type interaction parameter (18), χ, by using (19)

$$\chi = \beta + \frac{V}{RT}(\delta_S - \delta_P)^2 \quad (4)$$

where δ_P is the solubility parameter of the polymer, β is the lattice constant (whose value is generally taken to be equal to 0.34), RT is the energy term, and V is the molar volume of the solvent. The values of χ were calculated (20) using the predetermined values of δ_P, and these results are included in Table 4. The values of χ are higher by an order of magnitude for 1,2-dichloroethane and 1,1,1-trichloroethane, whereas lower values are observed for all the aromatic liquids. This further suggests that the interactions of haloalkanes are quite different than those of the aromatic liquids.

Both S and D values also increase with increasing temperatures. From the temperature dependence of diffusion and the sorption coefficients, the apparent activation energy values of diffusion (E_D) and sorption (ΔH_S) were calculated from the least-squares fit of the sorption and diffusion data according to the Arrhenius-type relation

$$X = X_0 \exp\left(\frac{-E_x}{RT}\right) \quad (5)$$

A typical Arrhenius plot for the dependence of ln D vs $1/T$ is shown in Fig. 2. The results of both E_D and ΔH_S, along with the estimated errors in ΔH_S, are presented in Table 4. The values of E_D for benzene (23.30 kJ·mol^{-1}) and methoxybenzene (36.30 kJ·mol^{-1}) are quite smaller than for the other liquids, for which E_D varied between 44.4 to 83.4 kJ·mol^{-1}. These values are comparable to those published in the literature for similar types of systems (16). The heat of sorption data, i.e., ΔH_S values, are positive in all cases, suggesting an endothermic type of sorption. However, the ΔH_S values for methoxybenzene (2.51 kJ·mol^{-1}) and toluene (5.3 kJ·mol^{-1}) are smaller than for the other liquids. The exceedingly high ΔH_S value of 47.8 kJ·mol^{-1} was found for nitrobenzene. These results can be further explained in terms of the Flory–Huggins theory (18). Sorption may thus be considered to be a combination of condensation and mixing phenomena, so that we can calculate heat of mixing, ΔH_{mix}, from the thermodynamic identity

$$\Delta H_{mix} = \Delta H_S + \Delta H_V \quad (6)$$

where ΔH_V is the heat of vaporization of the solvent molecules. The calculated values of ΔH_{mix} given in Table 4 indicate that a higher driving force is required for nitrobenzene than for the other liquids.

In order to find useful applications of polymer membranes in separation science, it is important to estimate the values of the solvent concentration

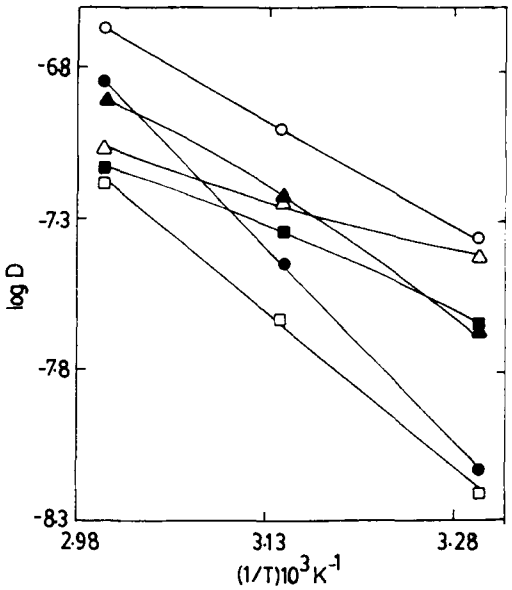

FIG. 2 Arrhenius plots of log D vs $1/T$ with the organic liquids given in Fig. 1.

profiles. Therefore, Eq. (1) was solved under the appropriate initial and boundary conditions to give (11, 12)

$$\frac{C_{(t,x)}}{C_\infty} = 1 - \frac{4}{\pi}\sum_{m=0}^{\infty}\frac{1}{(2m+1)}\exp\left[-\frac{D(2m+1)^2\pi^2 t}{h^2}\right]\sin\left[\frac{(2m+1)\pi x}{h}\right] \tag{7}$$

where $C_{(t,x)}$ and C_∞ are solvent concentrations expressed in weight %

TABLE 4
Activation Energy of Diffusion, Enthalpy of Sorption, Enthalpy of Mixing, and Interaction Parameter (χ) for Fluoroelastomer Membrane with Aromatic Liquids

Liquids	E_D (kJ·mol^{-1})	ΔH_S (kJ·mol^{-1})	ΔH_{mix} (kJ·mol^{-1})	χ
1,2-Dichloroethane	44.39 ± 0.36	9.10 ± 0.73	44.25	3.232
1,1,1-Trichloroethane	66.75 ± 4.13	11.04 ± 0.29	43.43	3.606
Benzene	23.30 ± 0.65	11.92 ± 2.77	45.76	0.366
Toluene	51.24 ± 5.00	5.26 ± 1.17	43.25	0.421
Methoxybenzene	36.30 ± 1.17	2.51 ± 0.73	49.35	0.340
Nitrobenzene	83.39 ± 2.25	47.80 ± 2.32	102.90	0.380

units at time t and distance x and at equilibrium sorption; m is an integer. Equation (7) was solved analytically using the values of D calculated from Eq. (2). The concentration profiles of liquids at different values of t and x have been calculated. Representative plots of these results for two liquids of widely differing diffusivities, viz., 1,1,1-trichloroethane ($D = 6.16 \times 10^{-9}$ cm^2·s^{-1}) and 1,2-dichloroethane ($D = 4.38 \times 10^{-8}$ cm^2·s^{-1}) at 30°C are displayed in Fig. 3. The effect of temperature on the shapes of such concentration profiles is presented in Fig. 4 for toluene at 30 and 60°C, respectively. From these plots it is realized that higher concentration profiles are observed for high diffusive liquids and vice versa.

The concentration profiles were also calculated numerically using a method based on the finite difference technique (12):

$$CN_m = \frac{1}{M}[C_{m-1} + (M - 2)C_m + C_{m+1}] \tag{8}$$

where the dimensionless parameter M is given as

$$M = \frac{(\Delta x)^2}{\Delta t}\frac{1}{D} \tag{9}$$

Here, C_m and CN_m are, respectively, the liquid concentrations at position m at time t and after the lapse of time Δt. For computational purposes,

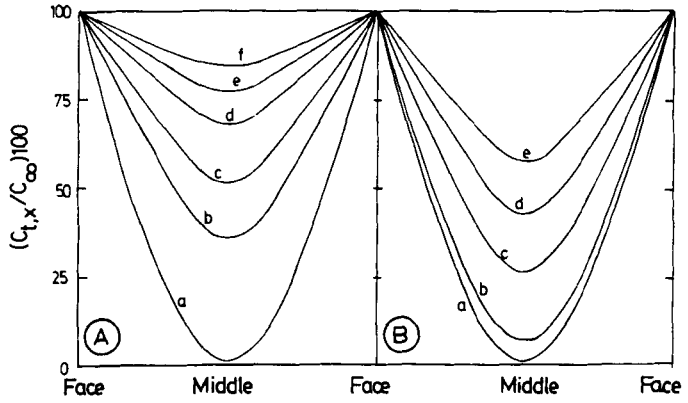

FIG. 3 Concentration profiles of (A) 1,2-dichloroethane with time intervals (in minutes) of a = 2.5, b = 50, c = 100, d = 250, e = 500, and f = 1000; and (B) 1,1,1-trichloroethane with time intervals (in minutes) of a = 70, b = 100, c = 250, d = 500, and e = 1000 at different penetration depths from one face to other of the polymer calculated from Eq. (7) at 30°C.

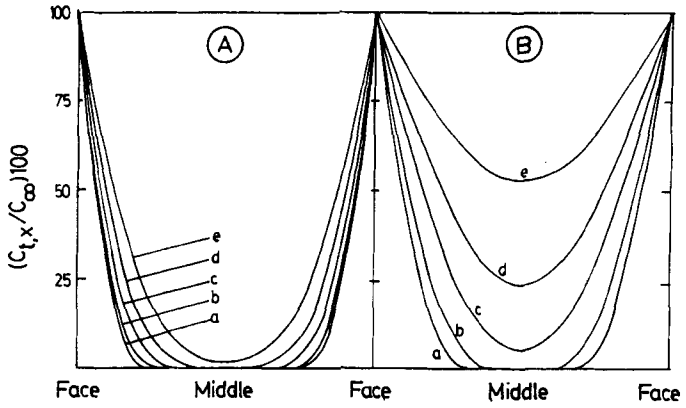

FIG. 4 Concentration profiles of toluene at (A) 30°C and (B) 60°C with time intervals (in minutes) of a = 50, b = 100, c = 250, d = 500, and e = 1000 at different penetration depths from one face to other of the polymer calculated from Eq. (8).

the membrane thickness is divided into 10 slices of equal size, Δx, and each slice is characterized by an integer, m. The liquid concentration profiles were calculated for each position for different time intervals by considering the unidirectional transport of liquids within the polymeric membrane material as shown in Fig. 5.

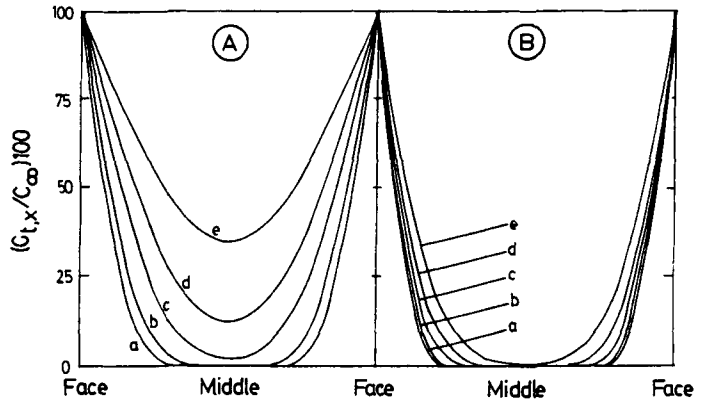

FIG. 5 Concentration profiles of (A) 1,2-dichloroethane and (B) 1,1,1-trichloroethane with time intervals (in minutes) of a = 50, b = 100, c = 250, d = 500, and e = 1000 at different penetration depths from one face to other of the polymer calculated from Eq. (8) at 30°C.

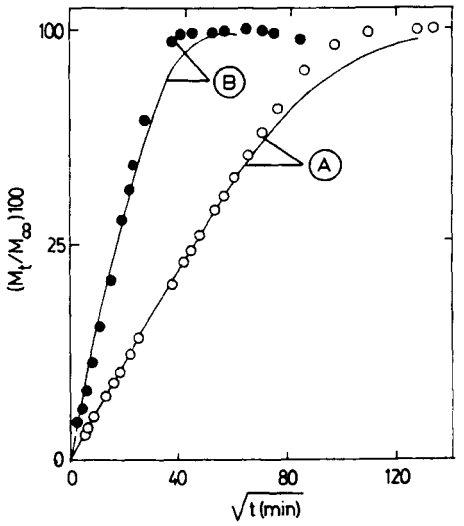

FIG. 6 Comparison of theoretical curves and the experimental sorption points of the polymer with toluene (A) at 30°C and (B) at 60°C.

In order to justify the accuracy of the diffusion coefficient values obtained from Eq. (2), the theoretical sorption curves were generated using (11)

$$\frac{M_t}{M_\infty} = 1 - \frac{8}{\pi^2} \sum_{m=0}^{\infty} \frac{1}{(2m+1)^2} \exp\left[- D \frac{(2m+1)^2 \pi^2 t}{h^2} \right] \qquad (10)$$

which was solved numerically for values of n from 0 to 10. The computed curves are compared in Fig. 6 with the experimental sorption points for toluene at 30 and 60°C. Good agreement between the theoretical curves and the experimental points is observed, further supporting the accuracy of the diffusivity results obtained from Eq. (2).

CONCLUSIONS

Knowledge of the sorption and diffusion parameters of liquid–polymer membrane systems is a key factor in deciding the end use of a polymeric membrane in MBSPs. Accurate estimation of these parameters is also helpful for a fundamental understanding of the transport phenomenon. In

the present paper, sorption and diffusion results of six organic liquids into the FC-2179 membrane are presented at 30, 45, and 60°C. Fick's diffusion equation was solved to calculate diffusion coefficients and concentration profiles. A numerical method was also used for the same purpose. However, the transport results of this study could not be explained satisfactorily in terms of the solubility parameter concepts of Hildebrand, and transport was found to be of the anomalous type. This study may have relevance in selecting suitable polymeric membranes in separation applications involving the contact of organic liquids. The polymer used was not attacked by any of the solvents used, and no extensive membrane swelling was observed in the presence of any of the liquids used.

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