



Organic solvents permeation through protective nitrile gloves

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Abstract

Effective diffusion coefficients for the permeation of several organic solvents through nitrile gloves were experimentally investigated using the American Society for Testing and Materials (ASTM) F-739 test cell method. Fick's effective diffusion coefficients for benzene, toluene, ethyl benzene, xylene, and styrene were estimated to be 0.61 ± 0.02 , 0.50 ± 0.06 , 0.27 ± 0.02 , 0.31 ± 0.03 , and 0.21 ± 0.03 ($\times 10^{-6}$ cm²/s), respectively. These results were comparable to results found by others using different permeation models. Using a transient mass diffusion equation and appropriate initial and boundary conditions, the effective diffusion coefficients were found to adequately simulate the concentration profiles of the organic solvents in the collection chamber during the permeation test. The effective diffusion coefficients were found to be inversely correlated to the molecular weight of the compounds. The results of this study can be used to estimate the exposure to workers using nitrile gloves.

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1. Introduction

Without personal protection equipment, the human skin is considered to be the primary defense in the workplace against exposure to hazardous materials such as organic solvents. Permeation of hazardous materials through the skin is one of the pathways of exposure that may result in skin diseases and occupational disorders. Gloves are one of the means of protection against exposure of hazardous materials during the handling of these materials. The impacts of the physical and chemical properties of polymeric gloves on permeation of

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organic solvents are of growing interest for the protection workers in contact with potentially hazardous chemicals.

To evaluate the effectiveness of polymeric gloves in protecting the users against hazardous chemicals, permeation test are generally conducted. Chemical permeation testing requires the challenge material or chemical to be placed in contact with the glove material. The chemical permeates through the glove material and is collected in an appropriate medium. The current method for the evaluation of the permeation of liquid chemicals is the American Society for Testing and Materials (ASTM) method F-739 [1]. Permeation involves the transport of chemicals through the material with diffusion as the dominant mass transfer mechanisms along with sorption and desorption [2].

To date, several researchers were concerned with the measurements of the steady state or maximum permeation rate and breakthrough detection time using the ASTM F-739 method. These two parameters provide information on the quantity of hazardous chemical and the time that the skin will be exposed to the chemicals through gloves. Gloves with low permeation rate may be considered a desirable barrier against the chemicals. However, these two permeation characteristics will vary with the type and thickness of glove materials, and the concentration of chemicals.

The permeation of chemical through a polymer film is usually described by Fick's Law. The permeation rate per unit area, J ($\text{ML}^{-2} \text{T}^{-1}$) is represented by:

$$J = -D \frac{dC_Z}{dZ} \quad (1)$$

where D is the Fick's Law diffusion coefficient of a chemical through the polymer film ($\text{L}^2 \text{T}^{-1}$); C_Z the concentration of chemical in the polymer film (ML^{-3}); and Z is the distance of permeation from the outside surface of polymer film (L). D is generally assumed to be a constant for different type of polymer material, and is neither a function of concentration C_Z nor distance Z [3,4].

According to Fick's second law, the concentration of the chemical, C_Z may be described by a one-dimensional diffusion equation:

$$\frac{\partial C_Z}{\partial t} = D \frac{\partial^2 C_Z}{\partial Z^2} \quad (2)$$

The concentration profile C_Z can be obtained by solving Eq. (2) with the necessary boundary and initial conditions. Table 1 summarizes the diffusion coefficients obtained by various studies for the permeation of organic solvent through polymeric film [3–10].

Table 1
Diffusion coefficients for organic solvents in polymeric film

References	Correlation	Parameters
[4,10]	$D = L^2/7.199t_{1/2}$	D : diffusion coefficient; L : thickness of membrane; $t_{1/2}$: time for the half-values of steady state permeation rate
[3,6,14]	$D = L^2/6t_1$	D : diffusion coefficient at large time; L : thickness of the polymer film; t_1 : lag time
[4,8,9]	$D = JL/S$	D : diffusion coefficient; L : thickness of polymer film; J : steady state permeation rate; S : solubility of chemical in polymer (determined by immersion test)

Crank used a diaphragm cell to estimate the diffusion coefficient of volatile organic compound vapors in polymer film [6]. To obtain solutions for Eq. (2), Crank assumed that (1) C_Z is a constant concentration on the surface ($Z = 0$) in contact with the chemical and is equal to the solubility of VOC vapor in the polymer film; (2) C_Z on the inside surface is assumed to be zero at all time. As shown in Table 1, the diffusion coefficient estimated by Crank was inversely dependent on the lag time through the membrane.

Similarly, Ziegel and his co-workers developed an empirical correlation to estimate the diffusion coefficient of solvents in polymer [8]. But instead of lag time, they used the time needed to achieve the half-values of steady state permeation rate. Based on Crank's assumptions, Vahdat indicated that D can be determined by Eq. (1) for solvent permeation through polymer film [4,8]. As presented in Table 1, the diffusion coefficient could be estimated using the steady state permeation rate and solubility S (ML^{-3}) of solvent in the polymer.

Vahdat's model has been widely applied to estimate the diffusion coefficient of solvent permeation through protective gloves. However, the solubility of solvent should be determined by a separate, long-term immersion experiment for several hours [4,8,11]. Glove samples were immersed in the chemical for a period of time, and the change of sample weight was measured before and after the immersion. Solubility was determined by the ratio of weight of chemical uptake in the sample over the volume of the dry sample.

In this study, permeation experiments using ASTM F-739 testing cell were conducted with several widely used organic solvents on nitrile gloves. Using the steady state permeation rate, the effective diffusion coefficients were estimated by using Fick's model. In addition, the effective diffusion coefficients were used to appropriately simulate the experimental permeation data. Finally, the correlations of effective diffusion coefficients were investigated using several physical and chemical properties of the organic solvents. The diffusion coefficient model of this research could be more applicable for the estimation of steady state permeation rate, and assessment of exposure for workers wearing the protective gloves.

2. Materials and methods

2.1. Organic solvents and nitrile gloves

The material tested was unlined (unsupported) nitrile glove and was obtained from Best Co. (Best 737–11, Menlo, GA). The thickness of the nitrile gloves, measured with a dial thickness gauge (Teclock Co., Japan), was 0.63 ± 0.03 mm. The specific gravity of the nitrile gloves was 0.92 ± 0.02 . Disks of approximately 8 cm in diameter were cut from the flat area of the nitrile gloves giving an approximate glove surface area of 20.8 cm^2 . The disks were rinsed with water, air-dried, and conditioned at 21 ± 5 °C and a relative humidity of $50 \pm 15\%$ for 24 h.

Organic solvents used for the experiments were benzene, toluene, ethyl benzene, xylene, and styrene. The physical–chemical properties of these five solvents are given in Table 2. The five organic solvents, purchased from Merck (Darmstadt, Germany) and Fluka (Switzerland), were of purity greater than 98%. The resistance of nitrile to permeation of these five organic compounds is recommended by its manufacturer. The solvents were chosen because

Table 2
Physical and chemical properties of organic solvents

	Solvent				
	Benzene	Toluene	Ethyl benzene	Xylene	Styrene
Grade	ACS Grade (Merck, 101783)	ACS Grade (Merck, 108325)	GC Grade (Fluka, 03082)	GC Grade (Merck, 108681)	GC Grade (Fluka, 85960)
Purity (%)	99	99	98	99.8	99.5
M_W^a	78.1	92.1	106.2	106.2	104.2
WS ^a	1780	515	187	198	300
K_{ow}^a	131.8	537.0	1412.5	1412.5	891.3
SG ^a	0.88	0.87	0.87	0.86	0.91
H^a	0.23	0.27	0.34	0.30	0.10
VIS ^a	0.73	0.55	0.60	0.70	0.72
S^b	1.51	1.27	1.01	0.99	2.19
LDL (ng) ^c	0.23	0.58	1.17	0.73	1.05

Properties are at 25 °C.

^a M_W , molecular weight; WS, water solubility in mg/l [15]; K_{ow} , octanol–water partition coefficient [15]; SG, specific gravity in kg/l; H , Henry's constant [15]; VIS, viscosity in centipoises [16].

^b S , solubility in nitrile, g/cm³ (determined by immersion experiment).

^c LDL, limit detection level.

of their wide use in industry and that they represent a wide range of water solubilities and Henry's law constants. As shown in Table 2, the water solubilities ranged from 187 mg/l for ethyl benzene to 1780 mg/l for benzene. Viscosities varied from 0.55 centipoise for toluene to 0.73 centipoise for benzene.

2.2. Permeation test

Permeation tests were performed according to the ASTM F739-96 chemical permeation method [1]. Fig. 1 shows the sketch of the open-loop test system and associated equipment. The disk sample was suspended in a glass test cell that separated the test cell into two hemispheres, the challenge chamber that contained the solvent and the collection chamber (volume of 100 ml) where the solvent was sampled and analyzed. The test cell has a diameter of 5.08 cm diameter and is made of glass (Pesce Laboratories, Kennett Square, PA).

The challenge chamber of the test cell was filled with 75 ml of the solvent covering the entire surface area of the disk sample. In the collection chamber, nitrogen gas at a flow rate of 125 ml/min was passed in contact with the glove material. The flow rate for nitrogen was controlled by a brass needle valve and was measured with a flow meter (Cole-Parmer, Part No. 12-10S, USA). An open-loop system as used in this study will minimize adsorption of organic solvent on the walls of the test cell.

The assembled test cell was immersed in a water bath maintained at a temperature of 25 ± 1 °C. To keep the nitrogen at the test temperature, nitrogen was passed through a glass chamber placed in the water bath prior to entering the test cell. All equipment was connected with Teflon tubing (Nalgene, USA) in the open-loop system.

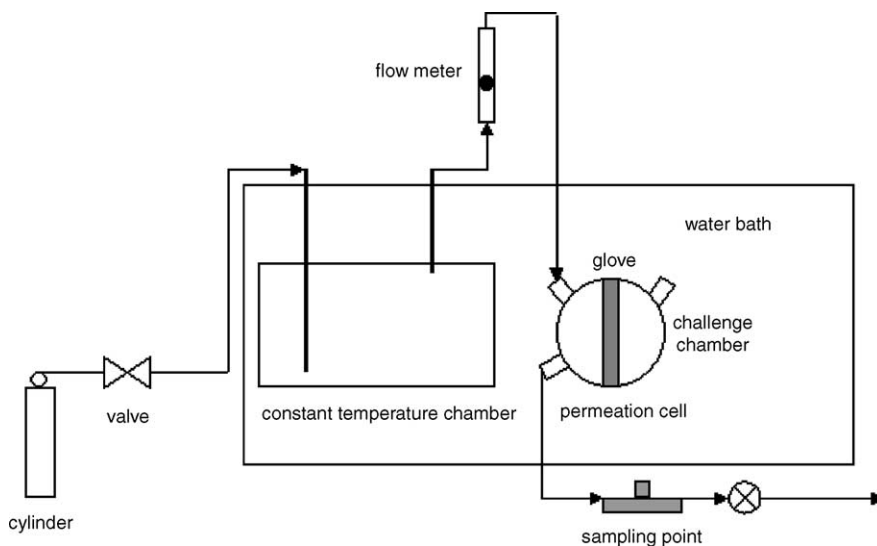


Fig. 1. Sketch of open-loop permeation cell system.

For each experimental run, 100 μl gas samples were collected from the sampling point every 10–15 min intervals using a gas-tight syringe after the solvent was added to the challenge chamber. The samples were immediately injected into a gas chromatograph equipped with a flame ionization detector (AutoSystem XL, Perkin-Elmer, USA). The experimental run was terminated when the solvent concentration in the collection chamber reached a steady state concentration. For each solvent, the experiment was repeated three times.

The oven temperature of the GC was set at an initial temperature of 60 $^{\circ}\text{C}$ for 1 min, then ramped to 150 $^{\circ}\text{C}$ at a rate of 8 $^{\circ}\text{C}/\text{min}$ and held for 1 min. The temperatures of the injection port and detector were maintained at 200 and 250 $^{\circ}\text{C}$, respectively. The column used was a DB-5 fused silica capillary column (30 m, 0.53 mm i.d., J&W). Nitrogen was used as carrier gas at a flow rate 30 ml/min. The correlation coefficients for all calibration curves were >0.995 . The limit detection levels (LDL) of five organic solvents are presented in Table 2.

2.3. Effective diffusion coefficients

The permeation rate J of mass transfer across the thin polymer film with thickness L may be represented as:

$$J = K(C^* - C) = D_e \frac{C^* - C}{L} = D_e \frac{C^*}{L} \quad (3)$$

where K is the mass transfer coefficient of solvent (LT^{-1}), and may be determined by the effective diffusion coefficient D_e (L^2T^{-1}) and L [4]; C^* the concentration of solvent in the challenge chamber, and is equal to the density ρ of chemical in this study (ML^{-3}); and C

is the concentration of chemical in the sampling chamber (ML^{-3}), and could be neglected as compared with the density ρ .

According to Eq. (3), the effective diffusion coefficient could be estimated with the steady state permeation rate [12]. As seen in Table 1, the diffusion coefficient estimated by Vahdat is similar to Eq. (3). But for Eq. (3), the concentration or density of chemical in the challenge chamber is used instead of the solubility S of the chemical in the material.

3. Results and discussion

3.1. Empirical diffusion coefficients

Fig. 2 shows the results of the experimental runs for all five solvents. As can be seen in Fig. 2, steady state concentrations were achieved within 3 h for all the solvents. At the end of the experimental run, the glove was found to increase in thickness by less than 10% and this change in thickness was assumed to be negligible. Using the steady state permeation rate for each solvent and Eq. (3), the Fick's effective diffusion coefficients (mean \pm S.D.) for benzene, toluene, ethyl benzene, xylene, and styrene were estimated to be 0.61 ± 0.02 , 0.50 ± 0.06 , 0.27 ± 0.02 , 0.31 ± 0.03 , and 0.21 ± 0.03 ($\times 10^{-6} \text{ cm}^2/\text{s}$), respectively.

In additional, 24 h immersion experiments were conducted for this study to obtain the solubility S of five organic compounds in nitrile. The solubilities, S ranged from 0.99 to 2.19 g/cm^3 that were greater than the density of the chemical (as seen in Table 2). Moreover, the solubility S was found to be significantly correlated to their density ($r = 0.976$, $P < 0.005$) and Henry's law constant ($r = 0.986$, $P < 0.005$). Table 3 shows the diffusion coefficients estimated by several models as given in Table 1 for the permeation experiments

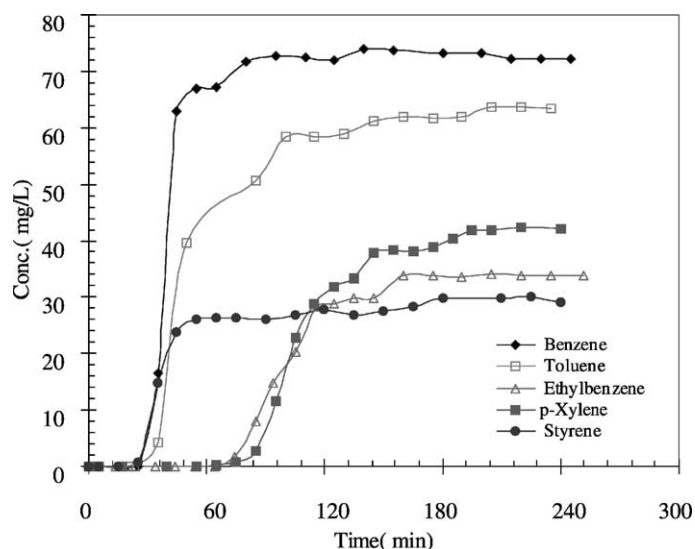


Fig. 2. Concentrations of five organic solvents in collection nitrogen.

Table 3
Comparisons of diffusion coefficients with literature model

	Solvent				
	Benzene	Toluene	Ethyl benzene	Xylene	Styrene
D_e	0.61 ± 0.02	0.50 ± 0.06	0.27 ± 0.02	0.31 ± 0.03	0.21 ± 0.03
D^\dagger	0.35 ± 0.01	0.34 ± 0.04	0.23 ± 0.02	0.27 ± 0.03	0.09 ± 0.01
D^\ddagger	0.35 ± 0.10	0.17 ± 0.03	0.12 ± 0.02	0.09 ± 0.01	0.25 ± 0.03
D^\S	0.24 ± 0.04	0.15 ± 0.02	0.10 ± 0.01	0.08 ± 0.01	0.23 ± 0.05

Mean \pm S.D. ($\times 10^{-6}$ cm²/s). D^\dagger : calculated by the equation $D = (J \times L)/S$ from Vahdat, 1987; D^\ddagger : calculated by the equation $D = L^2/6t_l$ from Crank, 1975; D^\S : calculated by the equation $D = L^2/7.199t_{1/2}$ from Ziegel, 1969.

of this study. The diffusion coefficients estimated using Crank's method were closed to the values estimated by Ziegel's equation. In general, the effective diffusion coefficients estimated by Eq. (3) were slightly higher than the values estimated by the other three methods.

3.2. Simulation of permeation experiments

The boundary and initial conditions for Eq. (2) for the experimental runs may be assumed as follows:

$$\begin{aligned} \text{BC : } & \begin{cases} C_Z(0, t) = C_0 = C^*, & t > 0 \\ C_Z(L, t) = 0, & t > 0 \end{cases} \\ \text{IC : } & C_Z(Z, 0) = 0 \end{aligned}$$

The solution for Eq. (2) with the above boundary and initial conditions is given as follows:

$$C_Z(Z, t) = C^* - C^* \left(\frac{Z}{L} \right) - \sum_{n=1}^{\infty} \left[\frac{2}{n\pi} C^* e^{-D(n\pi/L)^2 t} \sin \left(\frac{n\pi}{L} Z \right) \right] \quad (4)$$

In addition, the mass balance equation for the sampling chamber may be written as follows:

$$V \left(\frac{dC}{dt} \right) = -AD \frac{\partial C_Z(Z, t)}{\partial Z} \Big|_{Z=L} - QC \quad (5)$$

where V is the volume of sampling chamber; Q the flow rate of collection media nitrogen; A the surface area of glove exposed to the permeant. With an initial condition of $C(0) = 0$, the analytical solution for Eq. (5) is:

$$C = e^{-Q/vt} \frac{AD}{V} \left[\int^t \left(\frac{C_0}{L} + \sum_{n=1}^{\infty} \frac{2C_0}{L} e^{-D(n\pi/L)^2 t} \right) e^{Q/vt} dt \right] \quad (6)$$

The concentration with respect to time in Eq. (6) was solved using Maple-V software (Waterloo Maple Inc.). Fig. 3 shows the simulated concentration profile for styrene by using diffusion coefficients estimated from Eq. (3) and the experimental results, also plotted are

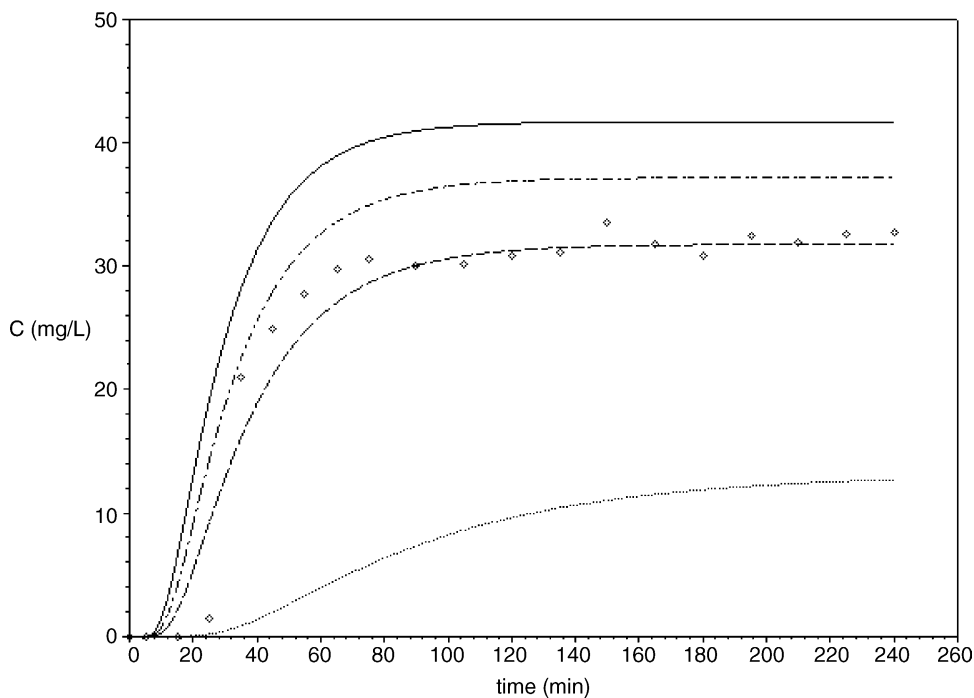


Fig. 3. Modeling results of styrene concentrations in collection nitrogen (curves from the upper using D^{\ddagger} , D^{\S} , D_e and D^{\dagger}).

the concentrations simulated using the diffusion coefficients estimated by others. Overall, as shown in Fig. 3, the diffusion coefficient obtained from Eq. (3) was able to simulate the permeation results. Moreover, the simulation results were greater than the experimental concentrations during the initial half-hour. This slight deviation from Fickian mechanism may be due to the effect of sorption on the permeation. Similar results were also found for the other four organic solvents.

As described earlier, Vahdat and Crank assumed that the concentration C_0 , boundary condition, was equal to solubility S for Eq. (2). Therefore, another simulations of Eq. (6) were performed using $C_0 = S$. For the five organic solvents, the experimental results could not be properly predicted using the diffusion coefficients determined by the four different methods. For example, Fig. 4 shows the simulation results for styrene and indicates that the diffusion coefficient D^{\dagger} obtained from Vahdat's method can simulate the styrene concentration profiles only for the initial half-hour and steady state conditions. This is reasonable because of the following two reasons. First, in Vahdat's method the diffusion coefficient was determined using the steady state permeation rate. Secondly, the solubility S may account for the organic solvent sorption on the glove. Therefore, Figs. 3 and 4 imply that sorption may play a role as the organic solvents initially permeated through the nitrile glove. However, Fig. 4 shows that Vahdat's method could not sufficiently predict the permeation time to reach steady state condition for styrene.

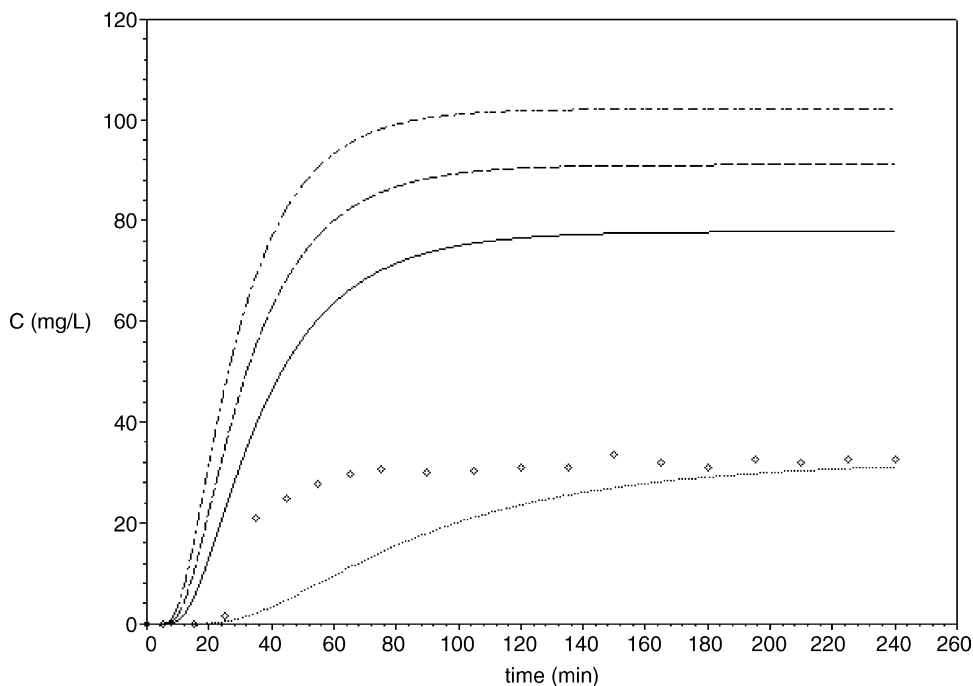


Fig. 4. Simulations of styrene concentrations in collection nitrogen using solubility for boundary conditions (curves from the upper using D^{\ddagger} , D^{\S} , D_e and D^{\dagger}).

Although the Crank's equation has been widely applied to determine the diffusion coefficients D^{\ddagger} for the permeation of liquid chemicals through polymer gloves [2,3,5,7], it is important to note that the Crank's equation is developed for solvent vapor permeation. The simulation results of Figs. 3 and 4 indicate that neither the solubility nor the concentration of solvent could be a suitable boundary condition in Eq. (2) for Crank's model. On the other hand, it is speculated that the immersion test may not be a proper estimation to determine the solubility of organic solvent in polymer.

Fig. 3 indicated that the effective diffusion coefficient and the boundary condition $C_0 = \rho$ for Eq. (2) could reasonably simulate the experimental permeation results. In comparison to Vahdat's method, the method used here does not require the solubility of the chemical in the material that was estimated by a separate immersion experiment. This will be advantageous for the application of effective diffusion coefficient to estimate the steady state permeation rate and to select protective gloves.

3.3. Correlation of diffusion coefficients in nitrile

Several researchers have provided correlations between the diffusion coefficient and the molecular weight (M_W), viscosity (VIS), and the octanol–water partition coefficient (K_{ow}) of chemical [2,4,7,9,13]. In general, chemicals with high molecular weights will have low

Table 4
Summary of diffusion coefficient correlation

Variable	Correlation	<i>r</i>	<i>P</i> -value
M_W	$D = 1.616 - 0.013M_W$	0.940	<0.05
WS	$D = 0.254 + 0.0002 WS$	0.840	>0.05
$\log K_{ow}$	$D = 1.327 - 0.337 \log K_{ow}$	0.870	>0.05
SG	$D = 2.737 - 2.688 SG$	0.312	>0.05
<i>H</i>	$D = 0.297 + 0.323H$	0.179	>0.05
VIS	$D = 0.504 - 0.192 VIS$	0.092	>0.05
$M_W, VIS, \log K_{ow}$ ^a	$D = 10^{37.63} M_W^{-22.714} VIS^{2.236} \log K_{ow}^{16.678}$	0.999	<0.05
M_W	$D = 10^{5.317} M_W^{-2.907}$	0.880	<0.05
VIS	$D = 10^{-0.578} VIS^{-0.655}$	0.185	>0.05
$\log K_{ow}$	$D = 10^{0.495} \log K_{ow}^{-2.136}$	0.796	>0.05

^a Stepwise.

diffusion coefficients. Table 4 shows that the effective diffusion coefficients estimated from this study were inversely correlated to molecular weight of organic solvents ($r = 0.94$, $P < 0.05$) while the viscosity was the least correlated ($r = 0.092$, $P > 0.05$). Zellers and Zhang also found that the diffusion coefficient was correlated to the molecular weight of chemical [9].

It has been found that the natural logarithm of diffusion coefficient in Viton gloves was significantly correlated to the natural logarithm of the solvent molecular weight [9,13]. Table 4 shows that the logarithm of D correlated well with the logarithm of the molecular weight ($r = 0.88$, $P < 0.05$). Vahdat investigated the diffusion coefficient of benzene, toluene and trichloroethane in nitrile, and found that the diffusion coefficients decreased linearly (on a log–log scale) with the viscosity of the chemical [4]. Their correlation was not significant in this study ($r = 0.185$, $P > 0.05$).

In addition, multiple regression analyses were performed using stepwise regression with 95% confidence limits. The best fitting correlation ($r = 0.999$, $P < 0.05$) was:

$$D = 10^{37.63} \frac{VIS^{2.236} \log K_{ow}^{16.678}}{M_W^{22.714}} \quad (7)$$

Several researchers have tried to correlate the diffusion coefficients to the breakthrough time for organic solvent permeation through nitrile and butyl gloves [7,14]. However, the breakthrough time of chemicals is sensitive to the detection limit of chemical analysis method. It should be emphasized that the correlation (Eq. 7) developed in this study is appropriate for nitrile gloves and may be different for the other materials.

4. Conclusion

Effective diffusion coefficients for various organic solvents in nitrile glove were estimated from experimental data. The effective diffusion coefficients for benzene, toluene, ethyl benzene, xylene, and styrene were estimated to be 0.61 ± 0.02 , 0.50 ± 0.06 , 0.27 ± 0.02 , 0.31 ± 0.03 , and 0.21 ± 0.03 ($\times 10^{-6} \text{ cm}^2/\text{s}$), respectively. The effective diffusion coefficient

coefficients estimated by this method were slightly higher than the values determined by other researchers. Instead of using the solubility of the chemical in the glove material which requires an additional experiment, this study used the density of the chemical in contact with the glove material. The estimated diffusion coefficients were found to correlate well with the molecular weight and were able to adequately simulate the concentration profiles of the actual experimental results. The simulation results indicated that the model developed in this study might be more feasible than the other approaches in the assessment of permeation of chemicals through nitrile gloves.

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