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Prediction of mutual diffusion coefficient in polymer solution

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Abstract

The solvent chemical potential derivative (i.e. $(\partial\mu_1/\partial\omega_1)_{T,P}$ is derived from the thermodynamic relation and mutual diffusion coefficients (D) are calculated with original UNIFAC-FV (Model 1) and the modified UNIFAC-FV (Model 2) without any assumption or simplification. Model 1&2 proposed in this work well correlate the experimental data of mutual diffusion coefficients in PIB/solvent systems and PMS-BR copolymer/solvent systems. It is shown that Model 1&2 are alternative tools to Flory–Huggins method for predicting mutual diffusion coefficients in polymer/solvent systems over wide ranges of temperature and concentration without any assumption. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Polymer/solvent systems; Mutual diffusion coefficient; UNIFAC-FV

1. Introduction

Polymer processing is often concerned with polymer solutions. As the transport phenomena of solvents in polymer solutions affect the polymerization, curing, polymer devolatilization, plasticization, pigment stability, preparation of polymer membranes, and other properties of polymer solution systems, the estimation method of solvent diffusion in polymer solutions has been extensively studied [1,2]. The solvent diffusion behavior in polymer matrix is also important to membrane separation processes such as pervaporation for the separation of volatile organic compounds (VOCs).

Reliable estimates of solvent diffusivity in polymer solutions has been required, and various diffusion models based on free-volume concept has been proposed [1,3,4]. Vrentas and Duda use the Flory–Huggins thermodynamic model in their free volume diffusion theory to describe the polymer-solvent enthalpic and entropic interactions. This model can describe athermal polymer/solvent systems fairly well [5]. For the estimates of solvent diffusion coefficient in polymer solution systems, free-volume parameters for the both polymer and solvent must be available [1,3,6–9]. The free-volume diffusion model developed by the Vrentas-Duda describes the solvent self-diffusion coefficient (D_1) and the polymer/solvent binary mutual diffusion coefficient (D) as given by Eqs. (1) and (2), respectively

$$D_1 = D_0 \exp\left(\frac{-E}{\mathrm{R}T}\right) \times \exp\left[\frac{-(\omega_1\hat{V}_1^* + \omega_2\xi\hat{V}_2^*)}{\omega_1\left(\frac{K_{11}}{\gamma}\right)(K_{21} - T_{g1} + T) + \omega_2\left(\frac{K_{12}}{\gamma}\right)(K_{22} - T_{g2} + T)}\right] \tag{1}$$

$$D = D_1(1-\phi_1)^2(1-2\chi\phi_1) \tag{2}$$

As shown above, Eq. (2) contains Flory–Huggins interaction parameter χ. χ is usually assumed to be constant, although it has been found to be a function of temperature and concentration. The derivative in Eq. (2) is based on χ being constant. The assumption of χ being constant is approximately correct for athermal systems where the enthalpy change on mixing is zero. The polymer/solvent interaction parameter has been written as a sum of entropic and enthalpic components. Both the enthalpic term and the free volume term are positive, but the enthalpic term decreases with temperature while the free volume term increases with temperature. χ should exhibit a minimum as a function of temperature due to these two competing effects [5]. Nakajima et al. [10] have shown that χ is concentration dependent for some poly(vinyl acetate) systems. Kokes et al. [11] have shown that χ is temperature dependent for poly(vinyl acetate) systems [5].

To predict D without any adjustable parameter from experiment, new prediction method has been needed. We

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Nomenclature

a_1	activity of solvent
b	geometric factor
c_1	external degrees of freedom per solvent molecule
D_i	self diffusion coefficient of component i (cm^2/s)
D_0	constant pre-exponential factor (cm^2/s)
E	critical energy which a molecule must possess to overcome the attractive forces holding it to its neighbors (cal/mol)
K_{11}	solvent free-volume parameter ($\mathrm{cm}^3/\mathrm{g/K}$)
K_{21}	solvent free-volume parameter (K)
K_{12}	polymer free-volume parameter ($\mathrm{cm}^3/\mathrm{g/K}$)
K_{22}	polymer free-volume parameter (K)
M	molecular weight of component i
M_{wm}	molecular weight of group m
r'_i, q'_i	pure component parameter
R	gas constant
R_k, Q_k	group parameter
Q'_k	group area parameter per gram
T	temperature (K)
$\hat{V}_i$	partial specific volume of component i
$\hat{V}_i^*$	specific critical hole free-volume of component i required for jump (cm^3/g)
W_m	weight fraction of group m in the mixture
z	coordination number
Greek letters	
θ'_m	area fraction of the group m
θ'_i	surface fraction of component i
Γ_k	group residual activity
$\Gamma_k^{(i)}$	group residual activity of group k in a reference solution containing only molecules of type i
v_i	volume per gram of component i
v_k^i	number of groups of type k in molecule i
$\tilde{v}_1$	reduced volume for the solvent
$\tilde{v}_M$	reduced volume for the mixture
ϕ'_i	segment fraction of component i
ϕ_i	volume fraction of component i
μ_i	chemical potential of component i
μ_i^0	chemical potential of pure i
χ	Flory–Huggins polymer/solvent interaction parameter
ρ_i	mass density of component i
ω_i	weight fraction of component i
ξ	ratio of critical molar volume of solvent jumping unit to that of polymer jumping unit
γ	overlap factor which is introduced because the same free volume is available to more than one molecule
Ψ_{mn}	group interaction parameter

can describe chemical potential term from the thermodynamic relation instead of Flory–Huggins and calculate with UNIFAC theories. It is known that UNIFAC, one of the group contribution methods, has lots of accumulated parameters and is widely used for estimating vapor–liquid equilibrium (VLE) and liquid–liquid equilibrium (LLE) system. In this work, we propose two models, which do not contain Flory–Huggins interaction parameter χ and do not include experimentally adjustable parameters. For the prediction of mutual diffusion coefficient (D), the derivatives of solvent chemical potential (i.e. $(\partial\mu_1/\partial\omega_1)_{T,P}$) were derived and calculated with original UNIFAC-FV (Model 1) and the modified UNIFAC-FV (Model 2) without any assumption or simplification. The results predicted by Model 1&2 were also compared with experimental data.

2. Theory

Since mutual diffusion coefficient (D) is useful for analyzing actual mass transfer phenomena, self-diffusion coefficient (D_1) must be related to the binary mutual diffusion coefficient (D). Vrentas-Duda [1] proposed the relationship between D and D_1 as follows:

$$D = \left(\frac{\rho_2\hat{V}_2\rho_1 D_1}{\mathrm{R}T}\right)\left(\frac{\partial\mu_1}{\partial\rho_1}\right)_{T,P} = \left(\frac{D_1 x_2}{\mathrm{R}T}\right)\left(\frac{\partial\mu_1}{\partial \ln x_1}\right)_{T,P} = \left(\frac{D_1\omega_1}{\mathrm{R}T}\right)(1-\omega_1)\left(\frac{\partial\mu_1}{\partial\omega_1}\right)_{T,P} \qquad (3)$$

For relating D_1 to D as shown in Eq. (3), derivative of solvent chemical potential (third term of the right hand side, i.e. $(\partial\mu_1/\partial\omega_1)_{T,P}$) must be obtained. The chemical potential derivative represents the effects of thermodynamic changes on the diffusion coefficient. Vrentas-Duda obtained the derivative of chemical potential (i.e. $(\partial\mu_1/\partial\omega_1)_{T,P}$) from the Flory–Huggins equation (Eq. (4)) under the assumption of constant χ

$$\mu_1 = \mu_1^0 + \mathrm{R}T[\ln(1-\phi_2) + \chi\phi_2^2 + \phi_2] \qquad (4)$$

The models proposed in this work, however, derive the chemical potential derivative (i.e. $(\partial\mu_1/\partial\omega_1)_{T,P}$) from the thermodynamic relation and calculate with original/modified UNIFAC-FV.

2.1. Model 1

As shown in Eq. (3), derivative of solvent chemical potential (i.e. $(\partial\mu_1/\partial\omega_1)_{T,P}$) is necessary to calculate mutual diffusion coefficient (D). Derivative of solvent chemical potential (i.e. $(\partial\mu_1/\partial\omega_1)_{T,P}$) is derived and calculated with original UNIFAC-FV instead of Flory–Huggins equation. Derivative of solvent chemical potential can be thermodynamically described with solvent activity as Eq. (5)

$$\left(\frac{\partial\mu_1}{\partial\omega_1}\right)_{T,P} = \mathrm{R}T\left(\frac{\partial \ln a_1}{\partial\omega_1}\right)_{T,P} \qquad (5)$$

Table 1
Parameter used in correlations of diffusion coefficients [7]

System	Parameter									
	$\hat{V}_1^*$ (cm^3/g)	$\hat{V}_2^*$ (cm^3/g)	K_{11}/γ (cm^3/g/K)	K_{12}/γ (cm^3/g/K)	$K_{21} - T_{g1}$ (K)	$K_{22} - T_{g2}$ (K)	χ	ξ	D_0 (cm^2/s)	E (cal/mol)
PIB/cyclohexane	1.008	1.005	3.02×10^{-3}	3.16×10^{-4}	−157.81	−117.93	0.55	0.44	1.04×10^{-3}	2630
PIB/*n*-pentane	1.158	1.005	2.41×10^{-3}	3.16×10^{-4}	−38.39	−117.93	0.77	0.43	7.32×10^{1}	9950
PIB/toluene	0.917	1.005	2.20×10^{-3}	3.16×10^{-4}	−102.72	−117.93	1.17	0.53	1.42×10^{-3}	1070
PIB/chloroform	0.510	1.005	7.12×10^{-4}	3.16×10^{-4}	−29.43	−117.93	1.09	0.35	9.02×10^{-3}	3780
PMS-BR(2%)/*n*-hexane	1.133	1.002	1.96×10^{-3}	3.13×10^{-4}	−41.08	−118.10	0.69	0.65	4.13×10^{-1}	4870
PMS-BR(7%)/*n*-pentane	1.158	0.994	2.41×10^{-3}	3.06×10^{-4}	−38.39	−119.39	0.60	0.40	3.72×10^{-2}	5280
PMS-BR(7%)/*n*-hexane	1.133	0.994	1.96×10^{-3}	3.06×10^{-4}	−41.08	−119.39	0.50	0.45	4.18×10^{-3}	3320
PMS-BR(15%)/*n*-pentane	1.158	0.981	2.41×10^{-3}	2.95×10^{-4}	−38.39	−120.56	0.62	0.46	2.49×10^{1}	9230
PMS-BR(15%)/*n*-hexane	1.133	0.981	1.96×10^{-3}	2.95×10^{-4}	−41.08	−120.56	0.71	0.64	3.56×10^{1}	7920
PMS-BR(15%)/cyclohexane	1.008	0.981	3.02×10^{-3}	2.95×10^{-4}	−157.81	−120.56	0.72	0.48	2.72×10^{-2}	4840

As the solvent activity in polymer solution [12] is expressed by

$$\ln a_1 = \ln a_1^{\mathrm{C}} + \ln a_1^{\mathrm{R}} + \ln a_1^{\mathrm{FV}} \tag{6}$$

We can obtain the derivative of solvent activity by differentiating Eq. (6)

$$\left(\frac{\partial \ln a_1}{\partial \omega_1}\right)_{T,P} = \left(\frac{\partial \ln a_1^{\mathrm{C}}}{\partial \omega_1}\right)_{T,P} + \left(\frac{\partial \ln a_1^{\mathrm{R}}}{\partial \omega_1}\right)_{T,P} + \left(\frac{\partial \ln a_1^{\mathrm{FV}}}{\partial \omega_1}\right)_{T,P} \tag{7}$$

Derivatives of combinatorial, residual, and free-volume terms in Eq. (7) result in the following expressions for solvent activity

$$\left(\frac{\partial \ln a_1^{\mathrm{C}}}{\partial \omega_1}\right)_{T,P} = \left(\frac{\partial \phi_1'}{\partial \omega_1}\right)_{T,P}\left(\frac{1}{\phi_1'} - 1\right) + \frac{z}{2}M_1 q_1'\left[\left(\frac{\partial \theta_1'}{\partial \omega_1}\right)_{T,P}\left(\frac{\theta_1' - \phi_1'}{\theta_1'^{2}}\right) + \left(\frac{\partial \phi_1'}{\partial \omega_1}\right)_{T,P}\left(\frac{1}{\theta_1'} - \frac{1}{\phi_1'}\right)\right] \tag{8}$$

where

$$\left(\frac{\partial \theta_1'}{\partial \omega_1}\right)_{T,P} = \frac{q_1' q_2'}{(q_1'\omega_1 + q_2'\omega_2)^2} \tag{9}$$

$$\left(\frac{\partial \phi_1'}{\partial \omega_1}\right)_{T,P} = \frac{r_1' r_2'}{(r_1'\omega_1 + r_2'\omega_2)^2} \tag{10}$$

$$\left(\frac{\partial \ln a_1^{\mathrm{R}}}{\partial \omega_1}\right)_{T,P} = \sum_k \nu_k^{(i)}\left[\left(\frac{\partial \ln \Gamma_k}{\partial \omega_1}\right)_{T,P} - \left(\frac{\partial \ln \Gamma_k^{(i)}}{\partial \omega_1}\right)_{T,P}\right] = \sum_k \nu_k^{(i)}\left[\left(\frac{\partial \ln \Gamma_k}{\partial \omega_1}\right)_{T,P}\right] \tag{11}$$

where

$$\left(\frac{\partial \ln \Gamma_k}{\partial \omega_1}\right)_{T,P} = M_k Q_k'\left[\frac{-1}{\sum_m \theta_m' \Psi_{mk}}\left\{\sum_m \Psi_{mk}\left(\frac{\partial \theta_m'}{\partial \omega_1}\right)_{T,P}\right\} - \sum_m \Psi_{km}\left\{\frac{\left(\frac{\partial \theta_m'}{\partial \omega_1}\right)_{T,P}}{\sum_n \theta_n' \Psi_{nm}} - \theta_m' \frac{\sum_n \Psi_{nm}\left(\frac{\partial \theta_n'}{\partial \omega_1}\right)_{T,P}}{\left(\sum_n \theta_n' \Psi_{nm}\right)^2}\right\}\right] \tag{12}$$

$$\left(\frac{\partial \theta_m'}{\partial \omega_1}\right)_{T,P} = Q_m'\left[\frac{\left(\frac{\partial W_m}{\partial \omega_1}\right)_{T,P}}{\sum_n Q_n' W_n} - W_m \frac{\sum_n Q_n'\left(\frac{\partial W_n}{\partial \omega_1}\right)_{T,P}}{\left(\sum_n Q_n' W_n\right)^2}\right] \tag{13}$$

$$\left(\frac{\partial W_{m,1}}{\partial \omega_1}\right)_{T,P} = \frac{G M_{wm} \nu_m}{M_{w1}} \tag{14}$$

$$\left(\frac{\partial W_{m,2}}{\partial \omega_1}\right)_{T,P} = -\frac{G M_{wm} \nu_m}{M_{w2}} \tag{15}$$

$$\left(\frac{\partial \ln a_1^{\mathrm{FV}}}{\partial \omega_1}\right)_{T,P} = C_1\left(\frac{\partial \tilde{v}_m}{\partial \omega_1}\right)_{T,P}\left[\frac{\tilde{v}_1}{\tilde{v}_m^2(1 - 1/\tilde{v}_1^{1/3})} - \frac{1}{(\tilde{v}_m - \tilde{v}_m^{2/3})}\right] \tag{16}$$

where,

$$\left(\frac{\partial \tilde{v}_m}{\partial \omega_1}\right)_{T,P} = \frac{v_1 r_2' - v_2 r_1'}{15.17b(r_1'\omega_1 + r_2'\omega_2)^2} \tag{17}$$

Substituting Eqs. (8), (11), and (16) into Eq. (7), derivative

Table 2
Comparison of calculated and experimental data

Solvent (1)	Polymer (2)	Temp. (°C)	Average error of *D* using original UNIFAC-FV (%)	Average error of *D* using modified UNIFAC-FV (%)	Average error of *D* using Flory–Huggins (%)	Remark
Cyclohexane	PIB	75	18.99	18.92	18.68	
		88	3.62	3.57	3.69	
		100	4.41	4.36	4.29	
n-Pentane	PIB	50	7.97	7.77	7.83	
		65	10.63	10.95	11.90	
		75	12.94	12.95	12.89	
Toluene	PIB	75	11.31	11.30	16.80	
		88	8.59	8.52	11.43	$\text{Error} = \frac{\lvert \text{exp.} - cal. \rvert}{\text{exp.}} \times 100$
		110	4.95	5.07	4.00	
Chloroform	PIB	75	0.98	0.85	2.16	
		88	3.39	3.65	4.08	
		100	2.48	2.82	3.38	
n-Hexane	PIB-BR (7%)	75	11.27	10.99	10.83	PIB: polyisobutylene
		100	2.14	2.73	4.32	
		125	12.28	11.96	11.24	
		175	9.43	9.59	9.71	
n-Pentane	PIB-BR (7%)	75	9.03	8.92	8.66	PMS: poly(p-methylstyrene
		100	3.36	3.39	3.42	
n-Hexane	PIB-BR (2%)	50	3.07	4.03	4.30	
		75	6.84	8.43	4.09	
n-Hexane	PIB-BR (15%)	50	6.49	18.74	6.09	
		75	10.53	13.89	5.38	Constant density of PMS
Cyclohexane	PIB-BR (15%)	50	8.73	8.69	9.14	
		75	19.82	20.69	17.99	
		100	11.42	11.84	7.77	
n-Pentane	PIB-BR (15%)	50	11.41	10.60	9.59	
		65	3.32	4.06	4.51	
		75	11.20	10.60	9.95	

of solvent chemical potential can be obtained. Therefore, we can calculate mutual diffusion coefficient (*D*) with the original UNIFAC-FV.

2.2. Model 2

UNIFAC-FV was modified by Larsen et al. [13], which is called modified UNIFAC-FV. Two changes are introduced in modified UNIFAC-FV: (1) the group-interaction parameters have been made temperature-dependent; and (2) the combinatorial term is slightly modified [13]. The modified UNIFAC-FV has the same form with original UNIFAC-FV except the combinatorial term. For the mutual diffusion coefficient of solvent, derivative of solvent chemical potential (i.e. $(\partial\mu_1/\partial\omega_1)_{T,P}$) was derived and calculated with the modified UNIFAC-FV instead of original UNIFAC-FV.

Combinatorial term is given by

$$\ln a_1^{\mathrm{C}} = \ln \phi_1' + 1 - \frac{\phi_1'}{x_1} = \ln \phi_1' + \phi_2' \tag{18}$$

where

$$\phi_i' = \frac{\omega_i r_i^{2/3}}{\sum_k \omega_k r_k^{2/3}} \qquad r_i = \frac{1}{M_i}\sum_k \nu_k^{(i)} R_k \tag{19}$$

Derivative of combinatorial term results in the following expression for solvent activity

$$\left(\frac{\partial \ln a_1^{\mathrm{C}}}{\partial \omega_1}\right)_{T,P} = \left(\frac{1}{\phi_1'} - 1\right)\left(\frac{\partial \phi_1'}{\partial \omega_1}\right)_{T,P} \tag{20}$$

where

$$\left(\frac{\partial \phi_1'}{\partial \omega_1}\right)_{T,P} = \frac{(r_1 r_2)^{2/3}}{(\omega_1 r_1^{2/3} + \omega_2 r_1^{2/3})^2} \tag{21}$$

Substituting Eqs. (8), (11), and (20) into Eq. (7), derivative of solvent chemical potential can be obtained. Therefore, we can calculate mutual diffusion coefficient (*D*) with the modified UNIFAC-FV.

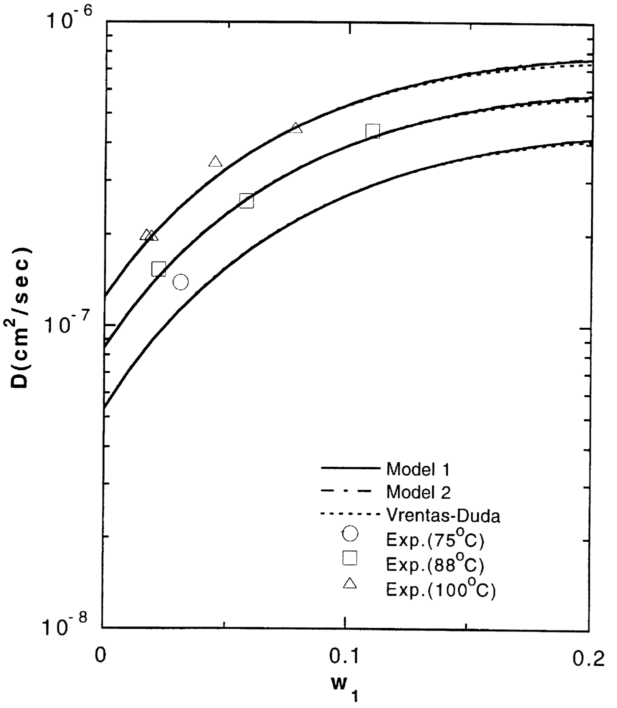

Fig. 1. Experimental data and theoretical correlations for PIB/cyclohexane mutual diffusion.

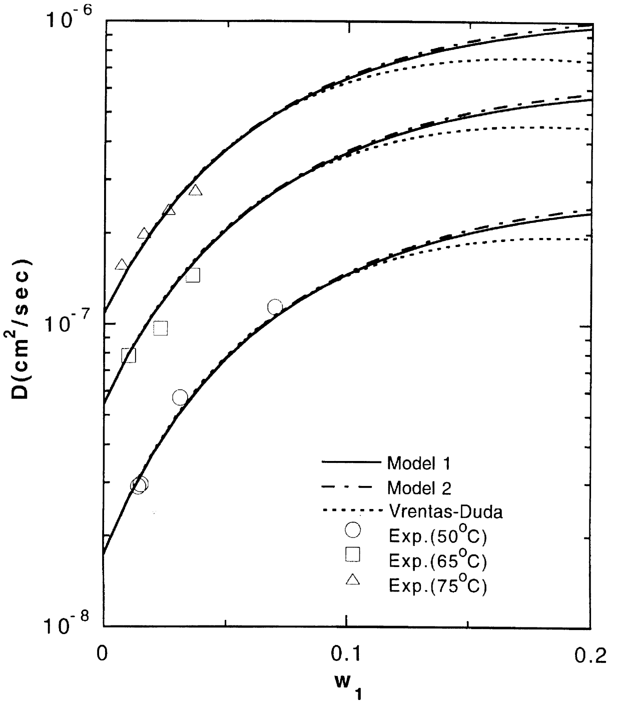

Fig. 2. Experimental data and theoretical correlations for PIB/*n*-pentane mutual diffusion.

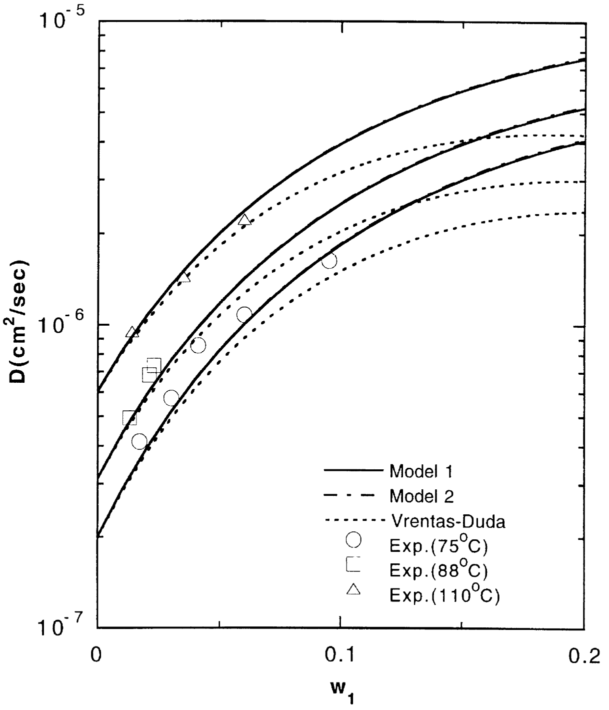

Fig. 3. Experimental data and theoretical correlations for PIB/toluene mutual diffusion.

3. Results and discussion

The mutual diffusion coefficients of polymer/solvent and copolymer/solvent system are obtained by Model 1&2 and Vrentas-Duda's. Parameters [7] used for the estimate of mutual diffusion coefficient are provided in Table 1. Predicted values and the experimental data [7] of mutual diffusion coefficient are provided and compared in Table 2.

3.1. PIB/solvent systems

Mutual diffusion coefficients (*D*) of PIB(polyisobutylene)/solvent (cyclohexane, *n*-pentane, toluene, chloroform) systems are represented in Figs. 1–4. For all the solvents investigated, the diffusion coefficient depends on both concentration and temperature. In rather higher range of solvent concentration, the values predicted by Model 1&2 are greater than those by Vrentas-Duda's. For the solvent of cyclohexane, *n*-pentane, and *n*-hexane, Model 1&2 proposed in this work well correlate with the experimental data at various temperatures and concentrations.

However, Model 1&2 predicted the experimental data better than Vrentas-Duda's for the PIB/toluene system. Since differences between *D* values predicted by Models 1& 2 and Flory–Huggins were mainly caused by solubility, differences from solubility estimation make differences of *D* values. Many researchers [14–16] reported that UNIFAC-FV is a good method to predict activity or activity coefficient of solvent such as *n*-hexane, *n*-pentane, toluene in PIB. When we add the average errors in the three columns in

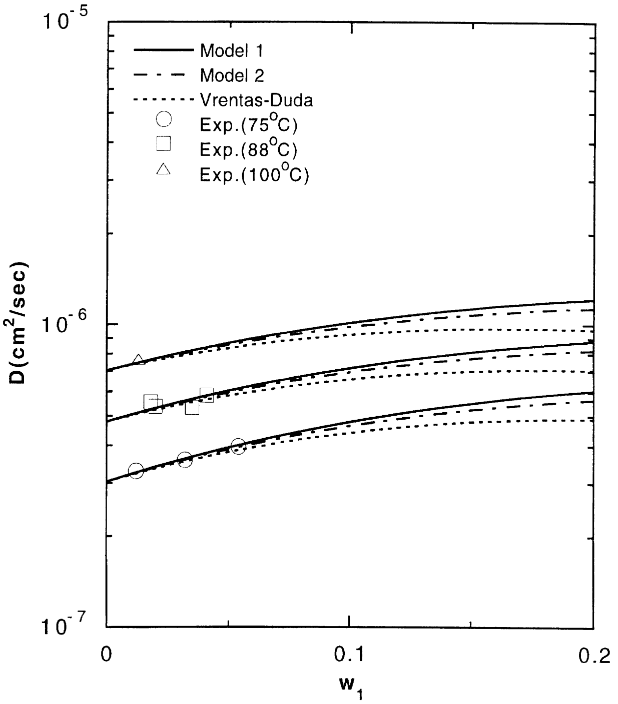

Fig. 4. Experimental data and theoretical correlations for PIB/chloroform mutual diffusion.

Table 2 and divide by the number of systems studied, we end up with 8.28% error for the original UNIFAC-FV, 8.30% for the modified UNIFAC-FV, and 10.74% for the Flory–Huggins. It seems that the solubility estimated by UNIFAC-FV is more accurate than those by Flory–Huggins relation in PIB/toluene system.

The values predicted by Model 1&2 for PIB/solvent systems are much comparable with each other.

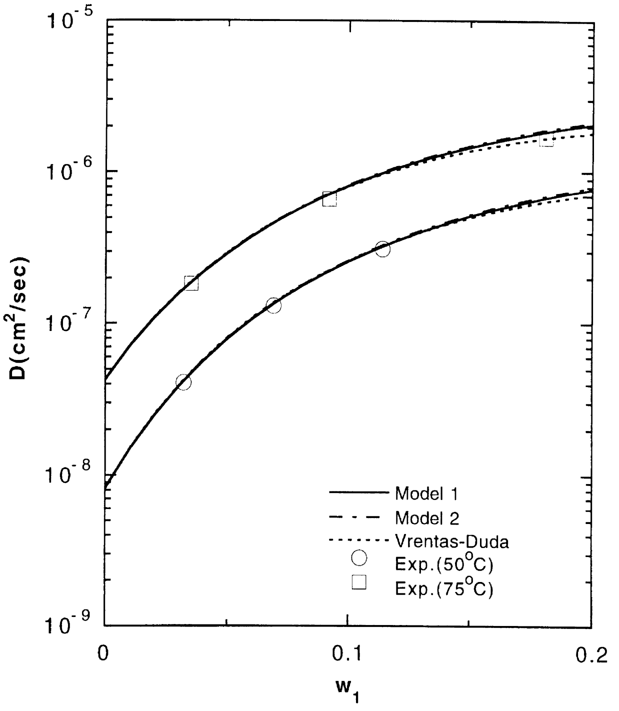

Fig. 5. Experimental data and theoretical correlations for PMS-BR(2%)/*n*-hexane mutual diffusion.

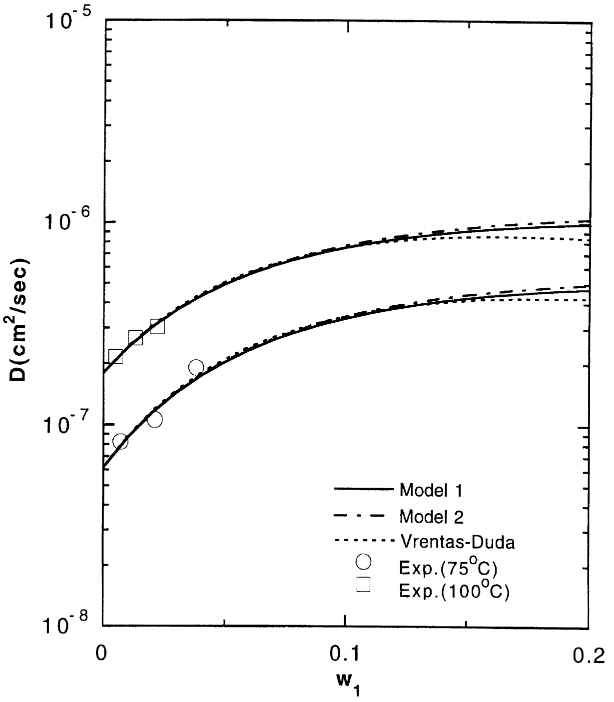

Fig. 6. Experimental data and theoretical correlations for PMS-BR(7%)/*n*-pentane mutual diffusion.

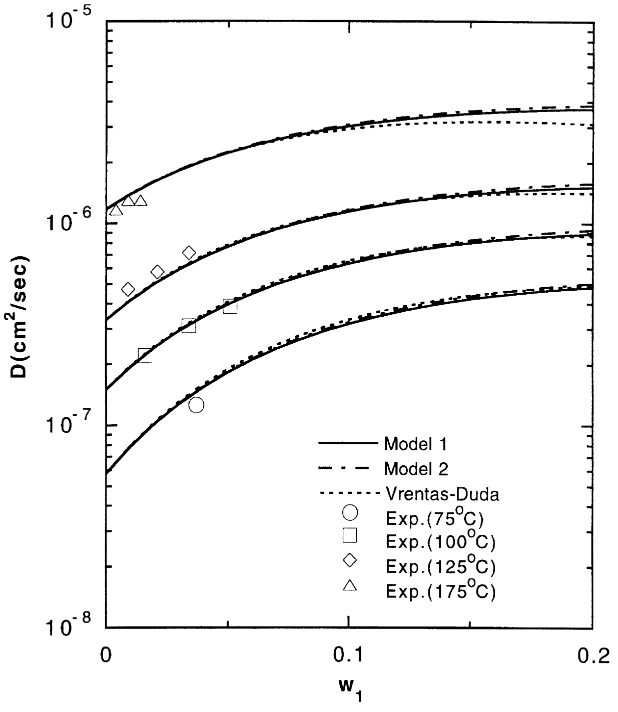

Fig. 7. Experimental data and theoretical correlations for PMS-BR(7%)/*n*-hexane mutual diffusion.

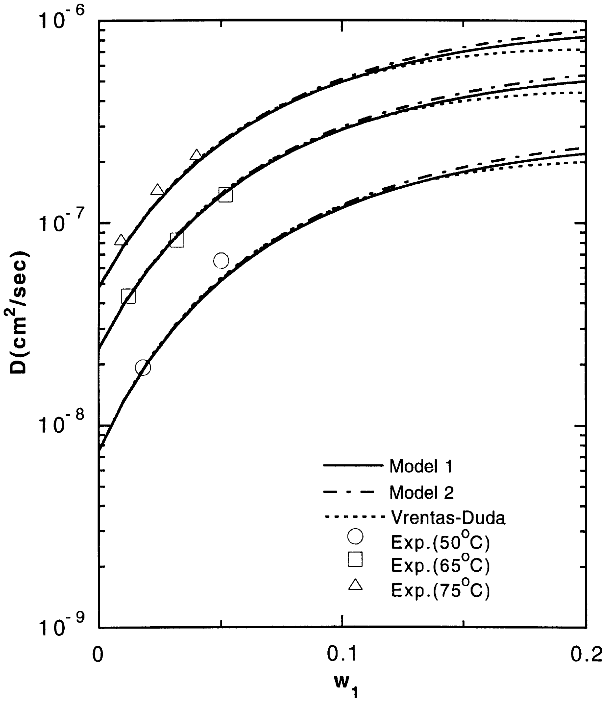

Fig. 8. Experimental data and theoretical correlations for PMS-BR(15%)/*n*-pentane mutual diffusion.

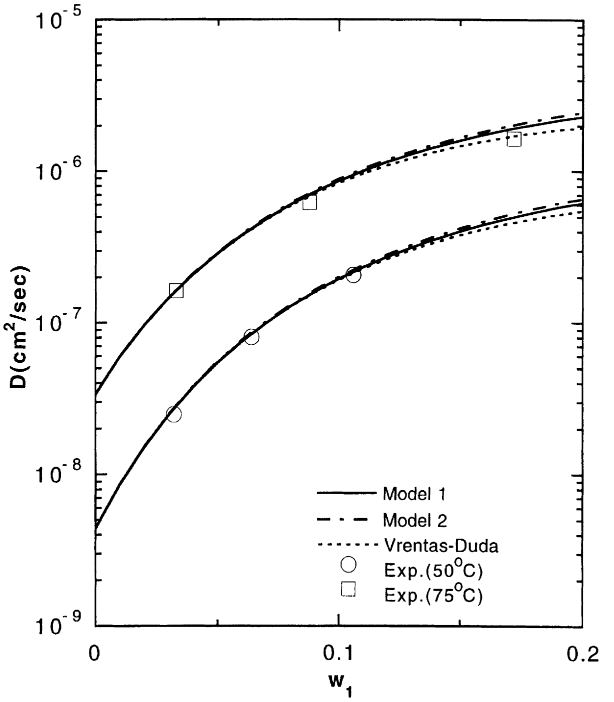

Fig. 9. Experimental data and theoretical correlations for PMS-BR(15%)/*n*-hexane mutual diffusion.

3.2. PMS-BR copolymer/solvent systems

Mutual diffusion coefficients (*D*) of PMS-BR copolymer/solvent system were predicted and compared with the experimental data for several organic solvents: *n*-hexane, *n*-pentane, and cyclohexane in PMS-BR (poly(*p*-methylstyrene)-*co*-isobutylene) containing 2, 7, 15 wt% of PMS, respectively. As shown in Figs. 5–10, Model 1&2 proposed in this work well correlate the experimental data at various temperatures and concentrations. In the rather higher range of solvent concentration, the predicted values by Model 1&2 are greater than those by Vrentas-Duda's free volume relation. The values predicted by Model 1&2 for PMS-BR copolymer/solvent system are very much the same with each other.

4. Conclusions

The solvent chemical potential derivative (i.e. $(\partial\mu_1/\partial\omega_1)_{T,P}$) are derived from the thermodynamic relation and mutual diffusion coefficients (*D*) are calculated with the original UNIFAC-FV (Model 1) and the modified UNIFAC-FV (Model 2) without any assumption or simplification. Model 1&2 well correlated the mutual diffusion coefficient data of cyclohexane, *n*-pentane, and *n*-hexane in PIB(polyisobutylene) and *n*-hexane, *n*-pentane, and cyclohexane in PMS-BR (poly(*p*-methylstyrene-*co*-isobutylene) systems. It is shown that Model 1&2 proposed in this work are alternative tools to Flory–Huggins theory for correlating and predicting mutual diffusion coefficients in polymer/solvent systems over wide ranges of temperature and concentration without any assumption or simplification. There is another point to be mentioned that mutual diffusion coefficients

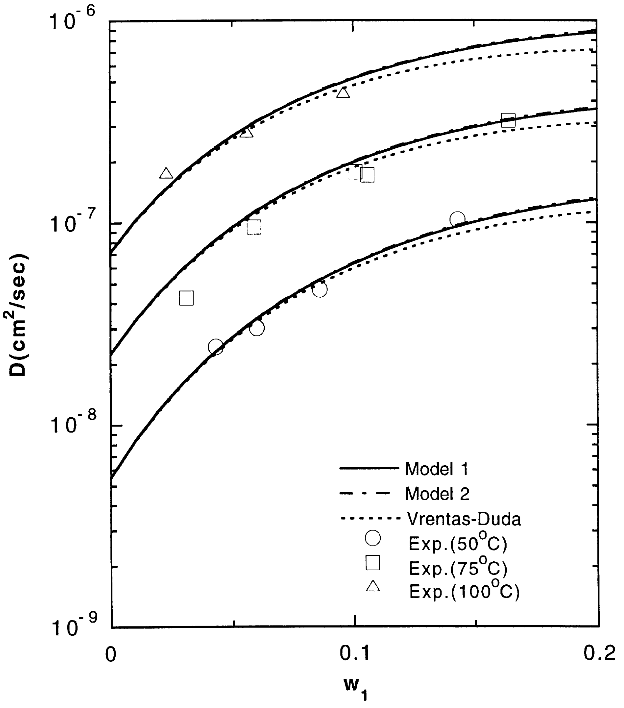

Fig. 10. Experimental data and theoretical correlations for PMS-BR(15%)/cyclohexane mutual diffusion.

calculated by Model 1&2 has little difference with each other in PIB/solvent and PMS-BR copolymer/solvent systems.

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