Diffusion Coefficients of Organic Compounds at Infinite Dilution in Mixtures Involving Associating Compounds. Experimental Determination and Modeling by Group Contribution Methods

Amor Safi, Christophe Nicolas, Evelyne Neau,* and Jean-Louis Chevalier

Laboratoire MSNM-GP, UMR 6181 Faculté des Sciences de Luminy, Université de la Méditerranée,
13288 Marseille, Cedex 9, France

Diffusion coefficients of aromatic compounds at infinite dilution in binary mixtures have been measured at 298.15 K with respect to composition using the Taylor dispersion technique. The binary mixtures considered in this work were from alcohols (methanol, ethanol) and acetone with hexane, heptane, cyclohexane, carbon tetrachloride, and chloroform; the aromatic compounds were benzene, benzaldehyde, acetophenone, and phenol. The purpose of this work was to study the influence of the polarity of the binary mixtures on diffusion coefficients. The data were correlated by the Leffler−Cullinan equation, using the measured values of diffusion coefficients in pure solvents and estimating the viscosity of the binary mixtures with group contribution methods. Special attention was paid to the capability of models to take into account the polarity of the mixtures; for this purpose, in addition to the Grunberg−Nissan model, the UNIFAC-VISCO group contribution method was also considered.

Introduction

This paper is a continuation of the study of the diffusion properties of several solutes in various binary mixture solvents at 298.15 K.[1,2] Experimental data of diffusion coefficients are useful for investigating the structure of liquids and improving molecular theories for the liquid state.[3] Such data are also useful for predicting the rate-limiting factor in chemical processes. Different methods were published in the literature for their measurement, such as the diaphragm cell,[4] the optical,[5–8] and the Taylor dispersion[9–11] techniques. The last one, which is one of the most widely used, was selected in this work as it is an accurate, rapid, and easy to handle method.[12–15]

Infinite dilution diffusion coefficients D_{1m}^{∞} of benzene were measured at 298.15 K in binary mixtures formed from alcohols (methanol, ethanol) and acetone with saturated components (hexane, heptane, cyclohexane, carbon tetrachloride, and chloroform). Three other solutes (benzaldehyde, acetophenone, and phenol) in methanol + ethanol mixed solvent were also measured.

Concerning the modeling of the experimental data, it was shown in previous papers[1,2] that the use of the experimental values D_{1p}^{∞} of solutes in pure solvents should always be taken into consideration. It was also observed that the Leffler−Cullinan[16] equation provides the best correlation of experimental diffusion coefficients assuming that the mixture viscosities can be predicted by means of reliable group contribution methods.

In a previous work devoted to the modeling of diffusion coefficients of aromatic components in alkane and cycloalkane solvents, satisfactory results were obtained by means of the Grunberg−Nissan[17] group contribution method. In this study, special attention was paid to the capability of models to take into account the polarity of the solvents. For this reason, in addition to the Grunberg−Nissan model, the UNIFAC-VISCO[18–20] group contribution method was also considered.

Experimental Section

Diffusion coefficients at infinite dilution were measured using the Taylor dispersion technique. The apparatus employed in this study is similar to the one used in our previous works[1,2] and described in the original publication of Huss et al.[21] In the present work, we will only recall the main features of the apparatus. It is based on the diffusive flow generated by the slow running of a solution, called carrier, in a long capillary tube and the dispersion of a very small amount of a solute injected into the carrier. A 655A liquid chromatographic pumping system (Merck-Hitachi, Tokyo, Japan) was used to provide the constant laminar flow, and the temperature was kept at 298.15 K using a thermostatted bath (Karlsruhe, F.R.G.). The detector was a Merck UV/vis filter photometer operating at 254 nm. The purity of reagents was superior to 99.9 %.

As previously reported,[1,21] the main sources of uncertainties in the calculation of the diffusion coefficients are the measurement of the retention time (0.5 %) and the determination of the width of the peak (1 %). Taking into account these experimental errors, the resulting uncertainty on the diffusion coefficients was estimated to be about 3 %.

The experimental values of diffusion coefficients measured in pure solvents are reported in Table 1. When available, literature values are also reported. It can be seen, as in our previous work,[2] that our measurements are consistent with those data, since they are inside the combined uncertainties of the published data.

The infinite dilution diffusion coefficients of benzene in the binary mixtures measured at 298.15 K are reported in Table 2. The influence of the aromatic solute was also checked, and data measured for benzaldehyde, acetophenone, and phenol in the methanol + ethanol solvent are given in Table 3.

---

* Corresponding author. E-mail: neau@luminy.univ-mrs.fr. Fax: +33 491 82 91 52.

10.1021/je700539w CCC: $40.75

Table 1. Experimental Values of Infinite Dilution Diffusion Coefficients D_{1p}^{∞} at 298.15 K and Comparison with Literature Values

		$10^5 D_{1p}^{\infty}/\mathrm{cm^2 \cdot s^{-1}}$	
solute	solvent	this work	lit.
benzene	hexane	4.70	4.79,[16] 4.76,[21] 4.72[30]
benzene	heptane	3.75	3.40,[29] 3.87,[30] 3.53,[36a] 3.91[31]
benzene	cyclohexane	1.92	1.90,[21] 1.86,[36a] 1.90,[31] 1.95[16]
benzene	acetone	4.25	4.28,[16] 4.16[32]
benzene	methanol	2.66	2.66[33]
benzene	ethanol	1.88	1.81[29]
benzene	chloroform	2.89	2.88,[34] 2.35[29]
benzene	carbon tetrachloride	1.45	1.41[35]
benzaldehyde	methanol	1.84	1.79[21]
benzaldehyde	ethanol	1.02	–
acetophenone	methanol	1.64	–
acetophenone	ethanol	0.91	–
phenol	methanol	1.76	–
phenol	ethanol	1.01	–

[a] Measurements performed at 297 K.

Results and Discussion

Diffusion coefficient data D_{1m}^{∞} of solute 1 in the mixture 2 + 3 were correlated by means of the Leffler–Cullinan[16] equation, which is expressed for a binary mixture by the following relation

$$\ln(D_{1m}^{\infty}\eta_m) = x_2 \ln(D_{12}^{\infty}\eta_2) + x_3 \ln(D_{13}^{\infty}\eta_3), \quad x_3 \approx 1 - x_2 \tag{1}$$

where η_m, η_2, and η_3 are, respectively, the viscosities of the mixture and of the pure components 2 and 3.

In this work, the viscosity η_m was estimated according to two group contribution methods developed in the literature according to the Eyring theory.[22]

- The Grunberg–Nissan method[17] assumes that

$$\ln(\eta_m) = x_2 \ln(\eta_2) + x_3 \ln(\eta_3) + x_2 x_3 G_m \tag{2}$$

which, as was previously shown,[2] allows expressing eq 1 as

$$\ln(D_{1m}^{\infty}) = x_2 \ln(D_{12}^{\infty}) + x_3 \ln(D_{13}^{\infty}) - x_2 x_3 G_m \quad \text{with} \quad G_m = \Sigma\Delta_3 - \Sigma\Delta_2 + W \tag{3}$$

where $\Sigma\Delta_i$ and W are estimated from the group contributions proposed by the authors.

- The UNIFAC–VISCO method proposed by Chevalier et al.[18–20]

In this method, the viscosity η_m of the mixture is correlated to those of the pure components η_2 and η_3 by means of the following relation

$$\ln(V_m\eta_m) = x_2 \ln(V_2\eta_2) + x_3 \ln(V_3\eta_3) + \frac{\Delta^* G^{\mathrm{E}}}{RT}, \quad x_3 \approx 1 - x_2 \tag{4}$$

where V_m, V_2, and V_3 are, respectively, the molar volumes of the mixture and of the pure components 2 and 3, and $\Delta^* G^{\mathrm{E}}$ is the excess molar free energy of activation which is estimated according to the group contributions developed in the original works.[18–20]

In the same way as for the Grunberg–Nissan method, the diffusion coefficient at infinite dilution D_{1m}^{∞} can by expressed from eq 1 as

$$\ln(D_{1m}^{\infty}) = x_2 \ln\left(D_{12}^{\infty}\frac{V_m}{V_2}\right) + x_3 \ln\left(D_{13}^{\infty}\frac{V_m}{V_3}\right) - \frac{\Delta^* G^{\mathrm{E}}}{RT} \tag{5}$$

In the original works,[18–20] the molar volume V_{m} of the mixture was approximated by the ideal mixture volume V_m^{id}. As was described in previous papers[23–28] devoted to the measurement of the mixture densities, the excess volumes of these mixtures are very small so that this approximation will not affect the diffusion coefficient estimation.

The results of the two correlations are presented in Table 4. All calculations are performed by using in eqs 3 and 5 the experimental values of D_{12}^{∞} and D_{13}^{∞} given in Table 1. AAD is the average relative absolute deviation

$$\mathrm{AAD} = \frac{100}{n}\Sigma\left|\frac{D_{1m}^{\infty,\mathrm{exptl}} - D_{1m}^{\infty,\mathrm{calcd}}}{D_{1m}^{\infty,\mathrm{exptl}}}\right| \tag{6}$$

where n is the number of experimental points.

The correlation of diffusion coefficients D_{1m}^{∞} given by eqs 3 and 5 essentially differ in the models considered for the representation of the free energies G_m and $\Delta^* G^{\mathrm{E}}$ of the mixed solvents. Hence, to get a significant interpretation of the results obtained with the two models, the different mixed solvents were classified, in Table 4, according to their thermodynamic properties; in particular, since the components 2 and 3 of the different mixed solvents have similar sizes, we have focused on the molar mixing enthalpy H^{E}/x_2x_3.

The first group of solvents consists of one alcohol (methanol or ethanol) with a saturated component (hexane, heptane, cyclohexane, chloroform, or carbon tetrachloride). The mixing properties of these solvents are mainly governed by the breaking of the hydrogen bonds between alcohol molecules. As shown in Figure 1 for the systems hexane + ethanol and carbon tetrachloride + ethanol, the H^{E}/x_2x_3 curve is highly asymmetrical, with a maximum effect for the infinite dilution of the alcohol in the saturated component; for both solvents, the value of the molar enthalpy of mixing at infinite dilution of ethanol is about (22 to 25) $\mathrm{kJ \cdot mol^{-1}}$, which means the order of magnitude of the hydrogen bond enthalpy. Results presented in Table 4 show that both the Grunberg–Nissan and UNIFAC–VISCO models provide a poor representation of the diffusion coefficients of benzene at infinite dilution in these solvents. As is also illustrated in Figure 2, the UNIFAC–VISCO model gives the worst results.

The second group of solvents concerns methanol or ethanol with acetone. Figure 3 shows, for the acetone + methanol mixture, that the molar enthalpies of mixing at infinite dilution are rather small, and the H^{E}/x_2x_3 curve is quite symmetrical. Indeed, the breaking of hydrogen bonds between methanol molecules is partially compensated by the formation of new bonds between methanol and acetone molecules. Figure 4 presents the diffusion coefficient of benzene at infinite dilution in acetone + methanol. As for the first group of solvents, both models provide poor results, and the UNIFAC–VISCO also gives the worst correlation of D_{1m}^{∞}.

The third group contains mixtures of acetone with a saturated component. For the acetone + hexane mixture, Figure 3 shows that the molar enthalpies of mixing at infinite dilution are greater than for the acetone + methanol mixture and that, by comparison with the hexane + ethanol system (Figure 1), the H^{E}/x_2x_3 curve presents a very moderate asymmetry in the dilute range of acetone. These effects can be explained by the breaking of interactions between acetone molecules, whose energies are smaller than those of the hydrogen bonds in alcohols. For these solvents, the UNIFAC–VISCO model provides a better estima-

Table 2. Experimental Infinite Dilution Diffusion Coefficients of Benzene (1) in Various Mixtures[a]

	$10^5D^{\infty}_{1m}$		$10^5D^{\infty}_{1m}$		$10^5D^{\infty}_{1m}$
x_2	$\mathrm{cm^2 \cdot s^{-1}}$	x_2	$\mathrm{cm^2 \cdot s^{-1}}$	x_2	$\mathrm{cm^2 \cdot s^{-1}}$
Hexane (2) + Ethanol (3)		Heptane (2) + Ethanol (3)		Cyclohexane (2) + Ethanol (3)	
0.0000	1.88	0.0000	1.88	0.0000	1.88
0.2024	2.13	0.1000	2.11	0.1894	1.97
0.2864	2.25	0.1835	2.30	0.3539	1.99
0.3942	2.45	0.3908	2.73	0.4506	2.01
0.4739	2.65	0.4991	2.99	0.5626	2.04
0.6082	3.05	0.6068	3.23	0.6898	2.06
0.7083	3.43	0.8001	3.60	0.8348	2.06
0.7796	3.82	1.0000	3.75	1.0000	1.92
0.8913	4.35				
1.0000	4.70				
Chloroform (2) + Methanol (3)		Carbon Tetrachloride (2) + Ethanol (3)		Acetone (2) + Methanol (3)	
0.0000	2.66	0.0000	1.88	0.0000	2.66
0.1962	2.26	0.2035	1.72	0.1220	2.95
0.3340	2.24	0.2998	1.70	0.2099	3.21
0.4441	2.23	0.4042	1.67	0.3594	3.50
0.5340	2.27	0.5047	1.64	0.5372	3.71
0.6117	2.40	0.6052	1.65	0.7061	3.87
0.6609	2.45	0.7033	1.63	0.7900	3.96
0.7958	2.60	0.8067	1.59	0.8781	4.12
0.8844	2.83	0.8980	1.56	1.0000	4.25
1.0000	2.89	1.0000	1.45		
Acetone (2) + Ethanol (3)		Acetone (2) + Hexane (3)		Acetone (2) + Cyclohexane (3)	
0.0000	1.88	0.0000	4.70	0.0000	1.92
0.1163	2.27	0.1926	4.82	0.1900	2.46
0.2030	2.54	0.4004	4.75	0.3000	2.71
0.3396	2.93	0.5055	4.68	0.5033	3.21
0.4016	3.12	0.7667	4.50	0.7449	3.80
0.5058	3.41	0.8910	4.39	0.8777	4.10
0.5437	3.50	1.0000	4.25	1.0000	4.25
0.7084	3.90				
0.8097	4.16				
1.0000	4.25				
Acetone (2) + Carbon Tetrachloride (3)		Carbon Tetrachloride (2) + Cyclohexane (3)			
0.0000	1.45	0.0000	1.92		
0.2164	1.84	0.2047	1.89		
0.3132	2.06	0.3317	1.85		
0.4050	2.28	0.4330	1.81		
0.5146	2.51	0.5333	1.75		
0.5823	2.69	0.6298	1.69		
0.6717	2.95	0.7166	1.64		
0.8005	3.38	0.7959	1.59		
0.9073	3.80	1.0000	1.45		
1.0000	4.25				

[a] x_2 is the mole fraction of component (2) in the mixed solvent.

Table 3. Experimental Infinite Dilution Diffusion Coefficients of Aromatics (1) in Methanol (2) + Ethanol (3) Mixtures[a]

	$10^5D^{\infty}_{1m}/\mathrm{cm^2 \cdot s^{-1}}$		
x_2	benzaldehyde	acetophenone	phenol
0.0000	1.02	0.91	1.01
0.2269	1.19	1.06	1.18
0.3477	1.29	1.15	1.26
0.4596	1.38	1.23	1.34
0.5484	1.45	1.29	1.41
0.6232	1.51	1.35	1.46
0.7067	1.58	1.41	1.52
0.7959	1.65	1.47	1.59
0.8961	1.74	1.55	1.67
1.0000	1.84	1.64	1.76

[a] x_2 is the mole fraction of component (2) in the mixed solvent.

tion of D^{∞}_{1m} than the Grunberg–Nissan model. Except for the acetone + carbon tetrachloride solvent, quite satisfactory results can thus be obtained.

The last group corresponds to nearly ideal systems, since the mixtures contain similar molecules, regarding both the size and the mutual interactions between the pure compounds. In that case, the UNIFAC–VISCO model is highly superior to the Grunberg–Nissan method for predicting the diffusion coefficients of aromatics at infinite dilution.

Taking into account all above remarks, it appears that the UNIFAC–VISCO model is unable to represent the free energies Δ^*G^E of mixed solvents containing one alcohol. Results presented in Table 4 highlight the necessity of developing a more specific model for the modeling of diffusion coefficients of aromatics in nonideal mixed solvents. At the present time, a new UNIFAC group contribution method is under development. With respect to the UNIFAC–VISCO model, the main feature of the new model is that the group contributions are directly determined from the infinite dilution coefficients, as expressed by eq 5.

Conclusion

Diffusion coefficients at infinite dilution of several aromatics in binary ideal, nonideal, and associated binary mixed solvents were experimentally determined at 298.15 K using the Taylor dispersion technique. Experimental determinations were correlated with the Leffler–Cullinan equation taking into account,

Table 4. Average Absolute Percent Deviations AAD between Experimental and Calculated Diffusion Coefficients of Aromatics at Infinite Dilution in Binary Mixture Solvents[a]

	solvents	solutes	*n*	Grunberg−Nissan	UNIFAC−VISCO
(I)	hexane + ethanol	benzene	8	10.1	31.8
	heptane + ethanol	benzene	6	3.6	8.7
	cyclohexane + ethanol	benzene	6	2.9	5.6
	chloroform + methanol	benzene	8	11.8	16.2
	carbon tetrachloride + ethanol	benzene	8	8.4	78.3
(II)	acetone + methanol	benzene	7	6.5	24.7
	acetone + ethanol	benzene	8	2.0	18.3
(III)	acetone + hexane	benzene	5	8.7	0.5
	acetone + cyclohexane	benzene	5	1.5	1.9
	acetone + carbon tetrachloride	benzene	8	21.9	20.7
(IV)	methanol + ethanol	benzaldehyde	8	17.4	2.7
	methanol + ethanol	acetophenone	8	17.5	2.8
	methanol + ethanol	phenol	8	17.5	2.7
	carbon tetrachloride + cyclohexane	benzene	7	13.7	2.8
	mean values		100	10.8	16.8

[a] Comparison of results obtained with different group contribution methods for the calculation of the mixture viscosity.

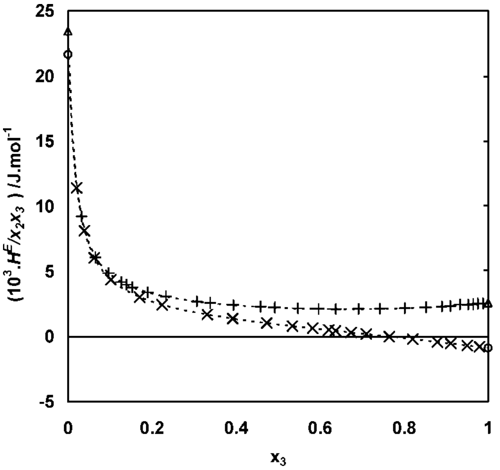

Figure 1. Experimental molar excess enthalpies H^{E}/x_2x_3 at 298.15 K for ethanol (3) with component (2): +, hexane;[37] ×, carbon tetrachloride.[38]

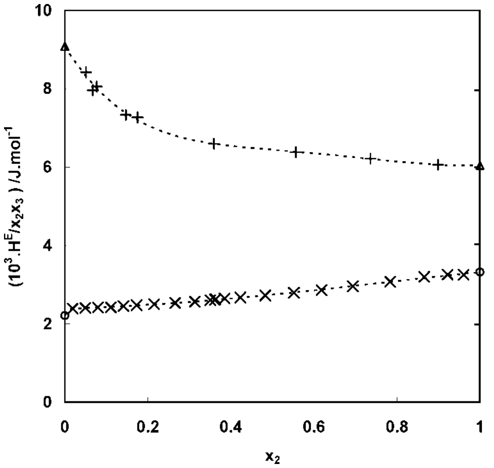

Figure 3. Experimental molar excess enthalpies H^{E}/x_2x_3 at 298.15 K for acetone (2) with component (3): +, hexane;[39] ×, methanol.[40]

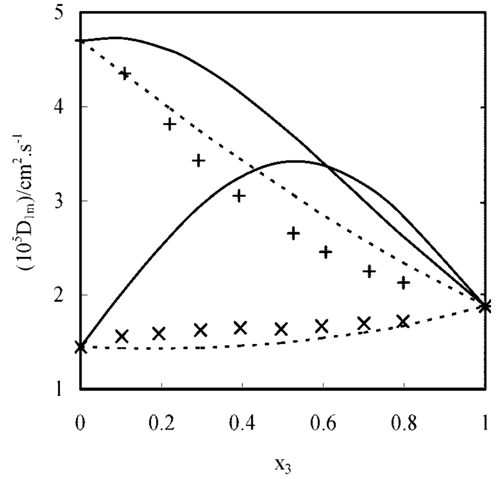

Figure 2. Correlation of experimental diffusion coefficients D_{1m}^{∞} of benzene (1) at infinite dilution in various mixed solvents of ethanol (3) with component (2): +, hexane; ×, carbon tetrachloride; using the Leffler−Cullinan equation. ——, UNIFAC−VISCO method; ----, Grunberg−Nissan method.

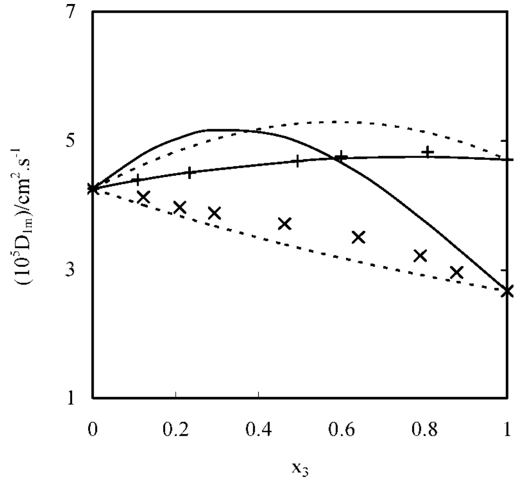

Figure 4. Correlation of experimental diffusion coefficients D_{1m}^{∞} of benzene (1) at infinite dilution in various mixed solvents of acetone (2) with component (3): +, hexane; ×, methanol; using the Leffler−Cullinan equation. ——, UNIFAC−VISCO method; ----, Grunberg−Nissan method.

as recommended in previous works,[1,2] the experimental values of diffusion coefficients in the pure solvents. Two group contribution methods were considered for the estimation of the mixture viscosity. Poor predictions of infinite dilution coefficients of aromatics in mixed solvents were obtained with the rough Grunberg−Nissan method. The second method, the UNIFAC−VISCO model, provides fair results for ideal systems or nonideal systems containing no alcohol. However, the model fails to represent diffusion coefficients in mixtures containing one alcohol with a saturated or polar component. The present study evidenced the need of a more general group contribution method for the representation of infinite dilution diffusion coefficients.

Literature Cited

(1) Safi, A.; Nicolas, C.; Neau, E.; Chevalier, J. L. Diffusion Coefficients of Aromatic Compounds at Infinite Dilution in Binary Mixtures at 298.15 K. *J. Eng. Chem. Data* **2007**, *52*, 126–130.

(2) Safi, A.; Nicolas, C.; Neau, E.; Chevalier, J. L. Measurement and Correlation of Diffusion Coefficients of Aromatic Compounds at Infinite Dilution in Alkane and Cycloalkane Solvents. *J. Chem. Eng. Data* **2007**, *52*, 977–981.

(3) Tyrrell, H. J. V.; Harris, K. R. *Diffusion in Liquids: A Theoretical and Experimental Study*; Butterworths: London, 1984.

(4) Stokes, R. H. An improved Diaphragm-cell for Diffusion Studies and some Tests of the method. *J. Am. Chem. Soc.* **1950**, *72*, 763–767.

(5) Kegeles, G.; Gosting, L. J. The Theory of an Interference Method for the Study of Diffusion. *J. Am. Chem. Soc.* **1947**, *69*, 2516–2523.

(6) Gosting, L. J. A Study of the Diffusion of Potassium Chloride in Water at 25° with the Gouy Interference Method. *J. Am. Chem. Soc.* **1950**, *72*, 4418–4422.

(7) Dunlop, P.; Gosting, L. J. A Study of the Diffusion of Glycolamide Chloride in Water at 25° with the Gouy Interference Method. *J. Am. Chem. Soc.* **1953**, *75*, 5073–5075.

(8) Thomas, W. J.; Furzer, I. A. Diffusion Measurements in Liquids by the Gouy Method. *Chem. Eng. Sci.* **1962**, *17*, 115–120.

(9) Taylor, G. I. Dispersion of Soluble Matter in Solvent Flowing Slowly Through a Tube. *Proc. R. Soc. A (London)* **1953**, *219*, 186–203.

(10) Taylor, G. I. Conditions Under Which Dispersion of Solute in a Stream of Solvent Can be used to Measure Molecular Diffusion. *Proc. R. Soc. A (London)* **1954**, *225*, 473–477.

(11) Aris, R. On the Dispersion of a Solute in a Fluid Flowing Through a Tube. *Proc. R. Soc. (London)* **1956**, *A235*, 67–77.

(12) Baldauf, W.; Knapp, H. Measurements of Diffusivities in Liquids by the Dispersion Method. *Chem. Eng. Sci.* **1983**, *38*, 1031–1037.

(13) Snijder, E. D.; te Riele, M. J. M.; Versteeg, G. F.; van Swaaij, W. P. M. Diffusion Coefficients of Several Aqueous Alkanolamine Solutions. *J. Chem. Eng. Data* **1993**, *38*, 475–480.

(14) Alizadeh, A.; Nieto de Castro, C. A.; Wakeham, W. A. The Theory of the Taylor Dispersion Technique for Liquid Diffusivity Measurements. *Int. J. Thermophys.* **1980**, *1*, 243–284.

(15) Alizadeh, A.; Wakeham, W. A. Mutual Diffusion Coefficients for Binary Mixtures of Normal Alkanes. *Int. J. Thermophys.* **1982**, *3*, 307–323.

(16) Leffler, J.; Cullinan, H. T. Variation of Liquid Diffusion Coefficients with Composition. *Ind. Eng. Chem. Fundam.* **1970**, *9*, 88–93.

(17) Grunberg, L.; Nissan, A. Mixture Law for Viscosity. *Nature* **1949**, *164*, 799–800.

(18) Chevalier, J. L.; Petrino, P.; Gaston-Bonhomme, Y. Estimation Method for the Kinematic Viscosity of a Liquid-Phase Mixture. *Chem. Eng. Sci.* **1988**, *43*, 1303–1309.

(19) Chevalier, J. L.; Petrino, P.; Gaston-Bonhomme, Y. Méthode d'estimation de la viscosité cinématique. *Entropie* **1986**, *129*, 35–42.

(20) Gaston-Bonhomme, Y.; Petrino, P.; Chevalier, J. L. UNIFAC-VISCO Group Contribution for Predicting Kinematic Viscosity: Extension and Temperature Dependence. *Chem. Eng. Sci.* **1994**, *11*, 1799–1806.

(21) Huss, V.; Chevalier, J. L.; Siouffi, A. M. Diffusion Coefficients in Water-Acetonitrile Mixtures. *J. Chromatogr.* **1989**, *500*, 241–255.

(22) Glasstone, S.; Laidler, K. J.; Eyring, H. *The Theory of Rate Processes;* McGraw-Hill: New York, 1941.

(23) Chien Wel, I.; Rowley, L. R. Binary Liquid Mixture Viscosities and Densities. *J. Chem. Eng. Data* **1984**, *29*, 332–335.

(24) Aminabhavi, V. A.; Aminabhavi, T. M; Balundgi, R. H. Evaluation of Excess Parameters from Densities and Viscosities of Binary Mixtures of Ethanol with, Anisole, N,N Dimethylformamide, Carbon Tetrachloride, and Acetophenone from 298.15 to 313.15 K. *Ind. Eng. Chem. Res.* **1990**, *29*, 2106–2111.

(25) Kouris, S.; Ponayiotou, C. Dynamic Viscosity of Mixtures of Benzene, Ethanol, and n-Heptane at 298.15 K. *J. Chem. Eng. Data* **1989**, *34*, 200–203.

(26) Noda, K.; Ohashi, M.; Ishida, K. Viscosities and Densities at 298.15 K for Mixtures of Methanol, Acetone, and Water. *J. Chem. Eng. Data* **1982**, *27*, 326–328.

(27) Papanastasiou, G. E.; Ziogas, I. I. Physical Behavior of Some Reaction Media. Density, Viscosity, Dielectric Constant, and Refractive Index Changes of Ethanol-Cyclohexane Mixture at Several Temperatures. *J. Chem. Eng. Data* **1991**, *36*, 46–51.

(28) Nigam, R. K.; Mahl, B. S. Strength of Interactions in Binary Mixtures of Methylene Chloride with Benzene, Toluene & Xylenes; Chloroform with Acetone, Ether & Dioxane; Benzene with Acetone, n-Pentane & n-Hexane; & of Cyclohexane with n-Pentane. *Indian J. Chem.* **1971**, *9*, 1255–1258.

(29) Reid, R. C.; Prausnitz, J. M.; Poling, B. E. *The Properties of Gases and Liquids*, 4th ed.; McGraw-Hill: New York, 1988.

(30) Harris, K. R.; Pua, C. K. N.; Dunlop, P. J. Tutual and tracer diffusion coefficients and frictional coefficients for the systems benzene-chlorobenzene, benzene-n-hexane, and benzene-n-heptane at 25. deg. *J. Phys. Chem.* **1970**, *74*, 3518–3529.

(31) Sanni, S. A.; Fell, C. J. D.; Hutchinson, H. P. Diffusion Coefficients and Densities for Binary Organic Liquid Mixtures. *J. Chem. Eng. Data* **1971**, *16*, 424–427.

(32) Anderson, D. K.; Hall, J. R.; Babb, A. L. Mutual Diffusion in Non-Ideal Binary Liquids Mixtures. *J. Phys. Chem.* **1958**, *62*, 404–409.

(33) Irani, R. R.; Adamson, A. W. Transport Processes in Liquid Systems. III. Thermodynamic Complications in the Testing of Existing Diffusional Theories. *J. Phys. Chem.* **1960**, *64*, 199–204.

(34) Sanni, S. A.; Hutchinson, P. Diffusivities and Densities for Binary Liquid Mixtures. *J. Chem. Eng. Data* **1973**, *18*, 317–322.

(35) Caldwell, C. S.; Babb, A. L. Diffusion in Ideal Binary Liquid Mixtures. *J. Phys. Chem.* **1956**, *60*, 51–56.

(36) Grushka, E.; Kikta, E. J. Extension of the Chromatographic Broadening Method of Measuring Diffusion Coefficients to Liquid Systems. Diffusion Coefficients of Some Alkylbenzenes in Chloroform. *J. Phys. Chem.* **1974**, *78*, 2297–2301.

(37) O'Shea, S. J.; Stokes, R. H. Activity Coefficients and Excess Partial Molar Enthalpies for (Ethanol + Hexane) from 283 To 318 K. *J. Chem. Thermodyn.* **1986**, *18*, 691–696.

(38) Nagata, I.; Tamura, K. Excess Molar Enthalpies of (Ethanol + Tetrachloromethane) and (Ethyl Ethanoate + Cyclohexane), and of (Ethanol + Benzene + Tetrachloromethane) and (Ethanol + Ethyl Ethanoate + Cyclohexane) at 298.15 K. *J. Chem. Thermodyn.* **1984**, *16*, 975–980.

(39) Murakami, S.; Amaya, K.; Fujishiro, R. Heat of Mixing for Binary Mixtures. The energy of Hydrogen Bonding between Alcohol and Ketone Molecules. *Bull. Chem. Soc. Jpn.* **1964**, *37*, 1776–1780.

(40) Nagata, I.; Tamura, K. Excess Enthalpies of Binary and Ternary Mixtures of Methanol with Acetone, Chloroform, Benzene, and Tetrachloromethane. *Fluid Phase Equilib.* **1983**, *15*, 67–69.

Received for review September 19, 2007. Accepted November 22, 2007.

JE700539W