

Excess molar volumes and viscosities of binary mixtures of 1,2-dimethoxyethane with chloroalkanes at 298.15 K

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MS received 1 February 1999; revised 24 March 1999

Abstract. Excess molar volumes (V_m^E), viscosity deviations ($\Delta\ln\eta$) and excess energies of activation for viscous flow (ΔG^{*E}) are reported for non-electrolyte mixtures of 1,2-dimethoxyethane (monoglyme) and dichloromethane, trichloromethane, and tetrachloromethane at 298.15 K and at atmospheric pressure over the whole mole fraction range. The Prigogine–Flory–Patterson (PFP) model has been used to calculate V_m^E , and the results are compared with experimental data. The Bloomfield and Dewan model has been used to calculate viscosity coefficients, which are compared with experimental data for three mixtures. These results have been analysed in terms of dipole–dipole interactions between 1,2-dimethoxyethane and chloroalkanes. The magnitude of the strength of interaction decreases with the dipole character of the molecule.

Keywords. Viscosity deviation; binary mixtures; 1,2-dimethoxyethane; chloroalkanes.

1. Introduction

Properties of liquid–liquid mixtures are technically very important as part of our studies on binary mixtures containing oxygen (–O–) and hydroxyl (–OH–) functional groups^{1–4}. We report here the excess molar volume (V_m^E), viscosity deviation ($\Delta\ln\eta$) and excess energies of activation for viscous flow (ΔG^{*E}) for mixtures containing 1,2-dimethoxyethane (DME) with dichloromethane (DCM), trichloromethane (TCM), and tetrachloromethane (TC) at 298.15 K and at atmospheric pressure, with the aim of characterizing their possible interactions. Excess properties of binary mixtures containing chloroalkanes have previously been studied by several authors^{5–8}. Excess thermodynamic properties of *n*-butoxyethanols with halogenated compounds of methane have also been determined in our laboratory^{1,9,10}. We are not aware of any study on the excess properties of binary mixtures of polyethers with chloroalkanes except with *bis*(2-methoxy)ether (diglyme) with chloroalkanes⁵.

Excess molar volumes and viscosities at different mole fraction have been measured for binary mixtures of polar molecules of different sizes and shapes. The V_m^E values have been analysed using the Prigogine–Flory–Patterson (PFP) model, which takes into account the hydrogen-bonded association.

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Viscosities of the mixtures, which are of general interest, have been analysed by means of the Bloomfield and Dewan method²⁹, based both on the free-volume and the absolute reaction rate theories.

2. Experimental

2.1 Materials

Materials were of the same origin and purity as used in earlier studies⁹. The sample 1,2-dimethoxyethane (Acros, USA, 99+%) was dried and fractionally distilled as described elsewhere¹⁰. All liquids were stored in dark bottles and carefully dried over 4 Å molecular sieves to reduce water contents. Before the actual measurements, all liquids were partially degassed under vacuum. Values of their densities recorded at 298.15 K and at atmospheric pressure (table 1) are in good agreement with those in the literature^{5,11-13}.

2.2 Apparatus and procedure

Excess molar volumes, reproducible to $\pm 0.003 \times 10^6 \text{ m}^3 \text{ mol}^{-1}$, were measured directly with a continuous dilution dilatometer as per Dickinson *et al.*¹⁴. Details of calibration, experimental setup and measuring procedure have been described elsewhere¹⁵. The mole fraction of each mixture was obtained to an accuracy of 1×10^{-4} from the measured apparent masses of the components. All apparent masses were corrected for buoyancy. Conversion to molar quantities was based on the relative atomic mass table of 1986 issued by IUPAC¹⁶. Each run covered just over half of the mole fraction range, giving an overlap between two runs.

The kinematic viscosities of pure liquids and liquid mixtures were measured at 298.15 K and at atmospheric pressure using an Ubbelohde suspended-level viscometer¹⁷. The viscometer was calibrated with thrice-distilled water and twice-distilled benzene. Care was taken to prevent evaporation during measurements. An average of four or five sets of flow times for each liquid were taken for the purpose of calculation of viscosity. Measured values of the kinematic viscosities were converted to dynamic viscosities (η) after multiplication by the density. The estimated error was $\pm 0.003 \text{ mPa.s}$. The flow-time measurements were made using an electronic stopwatch with a precision of $\pm 0.01 \text{ s}$. Densities of pure liquids were measured using a double-armed pycnometer with an

Table 1. Densities and viscosities of pure components at 298.15 K.

Substance	$\rho \text{ (g.cm}^{-3}\text{)}$		$\eta \text{ (mPa.s)}$	
	Exp.	Lit.	Exp.	Lit.
1,2-Dimethoxyethane	0.8626	0.86262 ¹¹ 0.8637 ¹² 0.86132 ¹³	0.420	0.424 ¹³
Dichloromethane	1.3165	1.3162 ⁵ 1.31678 ¹²	0.406	0.410 ⁵ 0.4043 ¹²
Trichloromethane	1.4794	1.4791 ⁵ 1.47970 ¹²	0.532	0.537 ⁵ 0.5357 ¹²
Tetrachloromethane	1.5843	1.5843 ⁵ 1.58436 ¹²	0.905	0.900 ⁵ 0.9004 ¹²

accuracy of 5 parts in 10^5 . The pycnometer was calibrated at 298.15 K with thrice-distilled water. A thermostatically controlled, well-stirred water-bath whose temperature was controlled to ± 0.01 K was used for all the measurements.

3. Results and discussion

Experimental results of the excess molar volumes for 1,2-dimethoxyethane + dichloromethane, trichloromethane and tetrachloromethane for a number of mole fractions at atmospheric pressure and 298.15 K are reported in table 2. A graphical representation of the experimental results together with the smoothing curves are shown in figure 1.

From the measured values of excess molar volumes, the densities of the liquid mixtures were calculated using the following equation:

$$\rho = (x_1M_1 + x_2M_2)/(V_m^E + x_1V_1 + x_2V_2), \quad (1)$$

where x_1 and x_2 are the mole fractions, M_1 and M_2 the molar masses, and V_1 and V_2 the molar volumes of 1,2-dimethoxyethane and chloroalkanes respectively. The accuracy in

Table 2. Excess molar volumes for 1,2-dimethoxyethane + chloroalkane systems at 298.15 K.

x_1	$V_m^E \times 10^6$ ($\text{m}^3 \cdot \text{mol}^{-1}$)	x_1	$V_m^E \times 10^6$ ($\text{m}^3 \cdot \text{mol}^{-1}$)	x_1	$V_m^E \times 10^6$ ($\text{m}^3 \cdot \text{mol}^{-1}$)
<i>1,2-Dimethoxyethane + dichloromethane</i>					
0.0254	-0.012	0.2655	-0.081	0.5919	-0.108
0.4568	-0.019	0.3131	-0.091	0.6625	-0.100
0.0725	-0.028	0.3408	-0.100	0.7445	-0.085
0.1040	-0.039	0.3702	-0.102	0.8352	-0.063
0.1412	-0.056	0.3760	-0.105	0.9111	-0.034
0.1713	-0.060	0.4078	-0.106	0.9633	-0.017
0.2067	-0.066	0.4156	-0.107		
0.2584	-0.080	0.5211	-0.107		
<i>1,2-Dimethoxyethane + trichloromethane</i>					
0.0266	-0.074	0.3214	-0.544	0.6944	-0.351
0.0549	-0.155	0.3450	-0.560	0.7398	-0.299
0.0751	-0.196	0.4120	-0.567	0.7819	-0.254
0.1217	-0.299	0.4864	-0.541	0.8752	-0.140
0.1633	-0.377	0.5372	-0.509	0.9315	-0.068
0.2353	-0.474	0.5967	-0.457	0.9693	-0.030
0.3040	-0.539	0.6505	-0.405		
<i>1,2-Dimethoxyethane + tetrachloromethane</i>					
0.0133	-0.004	0.3771	-0.062	0.6695	-0.051
0.0537	-0.012	0.4414	-0.065	0.7130	-0.054
0.1072	-0.023	0.4749	-0.069	0.7680	-0.049
0.2031	-0.039	0.5053	-0.069	0.8171	-0.041
0.2408	-0.046	0.5333	-0.070	0.8626	-0.036
0.2894	-0.051	0.5679	-0.068	0.9110	-0.025
0.3153	-0.056	0.6176	-0.063	0.9556	-0.012

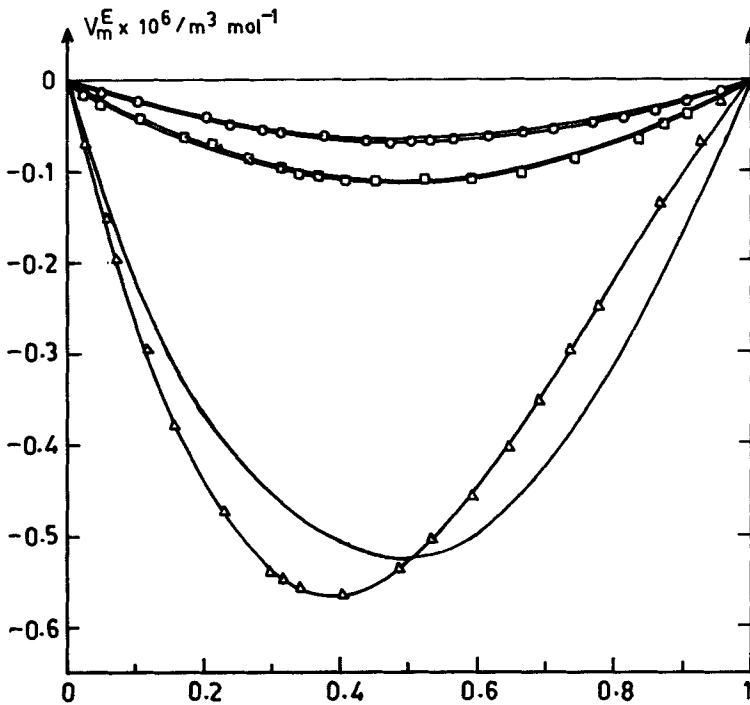


Figure 1. Experimental excess molar volumes for 1,2-dimethoxyethane + dichloromethane, \square ; + trichloromethane, Δ ; + tetrachloromethane, \circ . Continuous curves were calculated from (4) for experimental data and from PFP theory.

ρ due to the estimated accuracy in excess molar volume ($\pm 0.003 \times 10^6 \text{ m}^3 \text{ mol}^{-1}$) is $1 \times 10^{-1} \text{ kg m}^{-3}$.

The viscosity deviations were obtained from the following relationship^{18,19}

$$\Delta \ln \eta = \ln \eta - \sum_{i=1}^z x_i \ln \eta_i, \quad (2)$$

where η is the dynamic viscosity of the mixtures in mPa.s and η_i is the viscosity of the pure components respectively. The excess energies of activation for viscous flow were obtained using the expression

$$\Delta G^{*E} = RT \left\{ (\ln \eta V) - \sum_{i=1}^z (x_i \ln \eta_i V_i) \right\}, \quad (3)$$

where $V = \sum x_i M_i / \rho$. R and T have their usual meanings. Data on derived densities, viscosities and energies of activation for viscous flow for the three systems at 298.15 K are given in table 3.

Table 3. Densities, viscosities and excess energies of activation for viscous flow for the (1,2-dimethoxyethane + chloroalkanes) systems at 298.15 K.

x_1	ρ (g.cm ⁻³)	η (mPa.s)	ΔG^{*E} (J.mol ⁻¹)	x_1	$\rho \times 10^{-3}$ (g.cm ⁻³)	η (mPa.s)	ΔG^{*E} (J.mol ⁻¹)
<i>1,2-Dimethoxyethane + dichloromethane</i>							
0.0171	1.3042	0.423	104	0.4653	1.0524	0.466	373
0.0277	1.2967	0.425	116	0.5302	1.0244	0.464	354
0.0771	1.2630	0.434	179	0.5734	1.0067	0.462	337
0.1001	1.2479	0.438	210	0.6097	0.9924	0.461	324
0.1399	1.2227	0.445	253	0.6569	0.9744	0.456	290
0.2028	1.1852	0.454	310	0.6960	0.9600	0.453	267
0.2436	1.1621	0.457	329	0.7458	0.9424	0.448	228
0.2505	1.1583	0.461	347	0.8150	0.9190	0.439	162
0.3076	1.1279	0.464	367	0.8833	0.8973	0.431	96
0.3529	1.1050	0.466	379	0.9523	0.8764	0.423	30
0.4105	1.0773	0.468	385				
<i>1,2-Dimethoxyethane + trichloromethane</i>							
0.0221	1.4630	0.557	128	0.4359	1.1668	0.646	754
0.0489	1.4433	0.570	202	0.5060	1.1342	0.629	718
0.0775	1.4224	0.586	285	0.5591	1.1018	0.607	660
0.1176	1.3935	0.605	389	0.5993	1.0778	0.590	614
0.1721	1.3548	0.626	504	0.6486	1.0490	0.568	550
0.2143	1.3254	0.641	591	0.7139	1.0118	0.535	440
0.2690	1.2879	0.657	682	0.7540	0.9896	0.516	376
0.3051	1.2636	0.659	714	0.8174	0.9553	0.488	270
0.3442	1.2376	0.666	764	0.8600	0.9329	0.469	198
0.4078	1.1961	0.658	772	0.9322	0.8960	0.441	86
<i>1,2-Dimethoxyethane + tetrachloromethane</i>							
0.0177	1.5706	0.897	11	0.4983	1.2122	0.629	44
0.0514	1.5447	0.876	16	0.5447	1.1789	0.606	42
0.0797	1.5230	0.857	18	0.5969	1.1416	0.581	39
0.1051	1.5037	0.843	24	0.6426	1.1092	0.562	41
0.1496	1.4699	0.818	35	0.6955	1.0719	0.537	41
0.2080	1.4259	0.784	40	0.7380	1.0422	0.520	30
0.2738	1.3767	0.747	45	0.8135	0.9898	0.489	21
0.3409	1.3270	0.711	50	0.8911	0.9365	0.458	8
0.3806	1.2979	0.689	47	0.9584	0.8907	0.435	6
0.4421	1.2530	0.656	44				

The results of V_m^E , $\Delta \ln \eta$, and ΔG^{*E} are fitted to the Redlich-Kister type equation:

$$A = x_1 x_2 \sum_{i=1}^n a_i (x_1 - x_2)^{i-1}, \tag{4}$$

where a_i is the polynomial coefficient and n is the polynomial degree. The values of the coefficients, a_i , obtained by the least-squares method, with all points being given equal

Table 4. Coefficients a_i from (4) and standard deviation, σ , determined by the method of least squares.

		a_1	a_2	a_3	a_4	a_5	σ
1,2-Dimethoxyethane + dichloromethane	V_m^E	-0.439	-0.020	0.006			0.002
	$\Delta \ln \eta$	0.490	-0.166	-0.106	-0.317	0.372	0.007
	ΔG^{*E}	1484	-449	-259	-804	931	18
Trichloromethane	V_m^E	-2.134	1.144	0.191	-0.194		0.003
	$\Delta \ln \eta$	1.154	-0.607	-0.156			0.006
	ΔG^{*E}	2918	-1494	-970	70	1061	14
Tetrachloromethane	V_m^E	-0.270	-0.012	0.022	-0.043		0.002
	$\Delta \ln \eta$	0.073	-0.037	0.010			0.001
	ΔG^{*E}	180	-89	34			3

weightage, are presented in table 4 along with standard deviations, σ . In each case, the optimum number of coefficients was ascertained from an examination of the variation of the standard deviation with n :

$$\sigma = \left[\sum (A_{\text{cal}} - A_{\text{obs}})^2 / (n_{\text{obs}} - n) \right]^{1/2}, \quad (5)$$

where n_{obs} is the total number of direct experimental values. A represents V_m^E , $\Delta \ln \eta$ or ΔG^{*E} . For all the mixtures $\sigma(V_m^E) < 0.003$ for the precision attainable with the dilatometer used.

Excess volume versus composition plots in figure 1 show that the V_m^E values for all of the mixtures are negative over the whole mole fraction range. The negative values of V_m^E can be explained by considering the chemical or specific interactions which may have resulted from differences in molecular and free volumes²⁰ and possible dipole-dipole interactions leading to weak complex formation. Remarkably, V_m^E is most negative in the DME[1] + TCM[2] system. The reduction in the magnitude of V_m^E when dichloromethane is replaced by trichloromethane in the mixture is evidence for the occurrence of specific interaction between 1,2-dimethoxyethane and trichloromethane.

The algebraic values of V_m^E for all the three binary systems with 1,2-dimethoxyethane (monoglyme) are in the order TCM > DCM > TC. The behaviour is consistent with that of the V_m^E for bis(2-methoxy)ether (diglyme) with chloroalkanes⁵. This suggests that when the n -alkyl chain end of the polyether is fixed, V_m^E for both DCM and TC increases in magnitude in the positive direction; with each addition of a $-\text{OC}_2\text{H}_4-$ group in the monoglyme. However the V_m^E here is more negative for mixtures with TCM. The more significant decrease in V_m^E for the DME + TCM system indicates possible dipole-dipole interactions between the unlike molecules.

The Prigogine-Flory-Patterson (PFP) theory²⁰⁻²⁵ in the following form has been used to compute V_m^E of the mixtures:

$$\frac{V_m^E}{(x_1 V_1^* + x_2 V_2^*)} = \frac{(\bar{v}^{1/3} - 1)\bar{v}^{2/3}}{[(4/3)\bar{v}^{1/3} - 1]} \psi_1 \theta_2 \left(\frac{\chi_{12}}{\bar{v}} \right)$$

$$-\frac{(\bar{v}_1 - \bar{v}_2)^2[(14/9)\bar{v}^{-1/3} - 1]}{[(4/3)\bar{v}^{-1/3} - 1]} \psi_1 \psi_2 + \frac{(\bar{v}_1 - \bar{v}_2)^2(P_1^* - P_2^*)}{P_1^* \psi_2 + P_2^* \psi_1} \psi_1 \psi_2, \tag{6}$$

where ψ represents the contact energy fraction, given by

$$\psi_1 = 1 - \psi_2 = \frac{\varphi_1 P_1^*}{\varphi_1 P_1^* + \varphi_2 P_2^*}. \tag{7}$$

The values of the parameters for the pure liquid components required in the Prigogine-Flory-Patterson formalism derived using Flory's expression^{22,23} are presented in table 5. The χ_{12} parameter was adjusted by fitting the experimental V_m^E data over the whole concentration range. In table 5, the χ_{12} parameters obtained from the PFP theory was used to calculate V_m^E , and figure 1 gives the results together with the experimental values.

We have suggested before that the strong negative values of V_m^E of 1,2-dimethoxyethane (DME) + trichloromethane (TCM) system is the result of possible chemical interaction; the present results (again highly negative) for χ_{12} parameters (table 5) reinforce this suggestion.

In this work, dynamic viscosity has also been studied for these systems and often yields valuable information relating to the interaction between the molecules.

The positive values of $\Delta \ln \eta$ show that (i) the mixtures are more viscous than the corresponding ideal mixtures, and (ii) the specific interactions are present, as corroborated by the negative V_m^E and positive ΔG^{*E} values shown in figures 2 and 3. Positive values of the ΔG^{*E} parameter may be considered^{26,27} to be a useful indicator of the existence of specific interactions²⁸ between molecules. There are different expressions available in the literature to calculate η . Here we have applied the method of Bloomfield and Dewan²⁹.

Bloomfield and Dewan²⁹ developed an expression from the combination of the theories of free volume and absolute reaction rate,

$$\Delta \ln \eta = f(\bar{v}) - \Delta G^R / RT, \tag{8}$$

where $f(\bar{v})$ is the characteristic function of the free volumes defined by

$$f(\bar{v}) = 1 / (\bar{v} - 1) - x_1 / (\bar{v}_1 - 1) - x_2 / (\bar{v}_2 - 1), \tag{9}$$

Table 5. Parameters of pure components and Prigogine-Flory-Patterson theory (χ_{12}) at 298.15 K.

	$\alpha \times 10^{-4}$ (k ⁻¹)	$\kappa \times 10^7$ (k Pa ⁻¹)	$V^* \times 10^6$ (m ³ .mol ⁻¹)	$P^* \times 10^6$ (J.m ⁻³)	T^*	\bar{v}	\bar{T}	$\chi_{12} \times 10^6$ (J.m ⁻³)
1,2-Dimethoxyethane	12.77 ³²	11.15 ^a	80.25	579	4611	1.3019	0.0647	
Dichloromethane	13.91 ¹²	10.26 ¹²	48.77	707	4430	1.3227	0.0673	-11.7
Trichloromethane	12.60 ¹²	9.98 ¹²	62.13	635	4644	1.2987	0.0642	-36.4
Tetrachloromethane	12.29 ¹²	10.80 ¹²	10.80	567	4695	1.2928	0.0635	-4.2

^aCalculated by group additivity

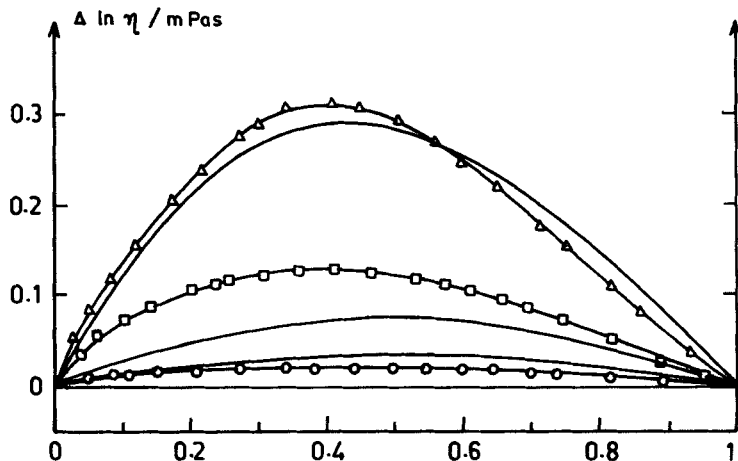


Figure 2. Experimental viscosity deviations for 1,2-dimethoxyethane + dichloromethane, \square ; + trichloromethane, Δ ; + tetrachloromethane, \circ . Continuous curves were calculated from (4) for experimental data and from Bloomfield and Dewan data²⁹.

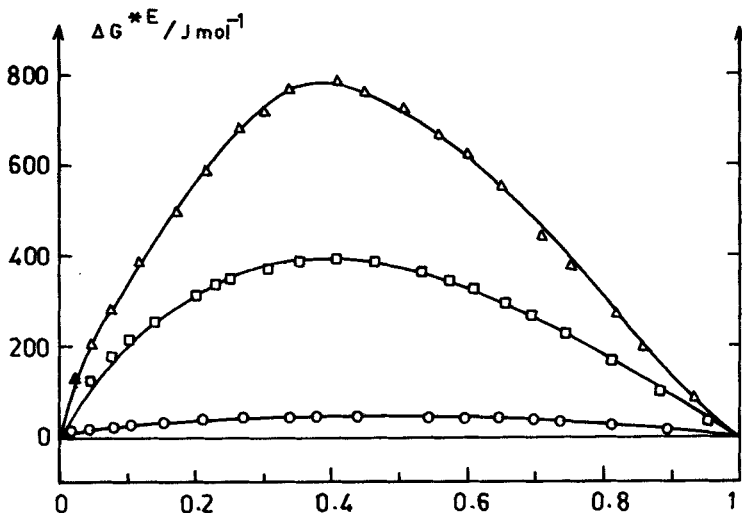


Figure 3. Excess energies of activation for viscous flow for 1,2-dimethoxyethane + dichloromethane, \square ; + trichloromethane, Δ ; + tetrachloromethane, \circ . Continuous curves were calculated from (4) for experimental data.

and ΔG^R is the residual energy of mixing, calculated using the expression³⁰

$$\Delta G^R = \Delta G^E + RT(x_1 \ln x_1/\phi_1 + x_2 \ln x_2/\phi_2). \quad (10)$$

The excess free energy can be obtained from the statistical theory of liquid mixtures proposed by Flory and co-workers and is given by

$$\Delta G^E = x_1 P_1^* V_1^* [1/(\bar{v}_1) - 1/(\bar{v}) + 3T_1 \ln\{(\bar{v}_1^{1/3} - 1)/(\bar{v}^{1/3} - 1)\}] \\ + x_2 P_2^* V_2^* [1/(\bar{v}_2) - 1/(\bar{v}) + 3T_2 \ln\{(\bar{v}_2^{1/3} - 1)/(\bar{v}^{1/3} - 1)\}] + (x_1 \theta_2 V_1^* \chi_{12}) / \bar{v}, \quad (11)$$

where the various symbols used have their usual meanings. A similar treatment was given by Katz *et al*⁵ and Kanti *et al*³¹.

Using the χ_{12} values from the fitting values of V_m^E and table 5, we have calculated ΔG^R and $f(\bar{v})$ and finally $\Delta \ln \eta$, according to the Bloomfield and Dewan relationship, which is compared with the experimental data (figure 2). Figure 2 shows that the largest deviation between the estimated and experimental curve occurs for 1,2-dimethoxyethane (DME) + dichloromethane (DCM).

It is interesting to note that in going from tetrachloromethane to trichloromethane, the absolute values of V_m^E become about five times larger. Since interaction between chloroalkanes and the ether DME cannot imply hydrogen bonding, the relatively larger value of the dipole moment of trichloromethane ($\mu = 1.15$ compared with $\mu = 1.14$ and 0 of dichloromethane and tetrachloromethane) may be responsible for a decrease in the negative contribution in V_m^E as is observed here.

Further, an analysis of each of the three contributions to the V_m^E , $\Delta \ln \eta$, and ΔG^{*E} of the present mixtures suggests the presence of dipole-dipole interactions in the DME + TCM mixture.

Acknowledgement

Financial support for this project from the Department of Science and Technology, New Delhi is gratefully acknowledged.

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