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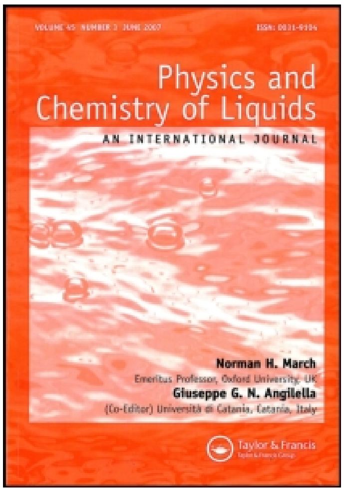

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Physico-chemical properties of binary mixtures of 1-butanol with chloroethanes or chloroethenes at 298.15 K

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Physico-chemical properties of binary mixtures of 1-butanol with chloroethanes or chloroethenes at 298.15 K

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The densities ρ, speeds of sound u, and viscosities η, of pure 1-butanol, 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2-tetrachloroethane, trichloroethylene, and tetrachloroethylene and those of their binary mixtures have been measured at 298.15 K and atmospheric pressure over the entire range of compositions. Excess molar volumes V^E, viscosity deviations $\Delta\eta$, deviation in compressibilities $\Delta\kappa_s$ and excess Gibbs energy of activation G^{*E}, were obtained from the experimental results and those were fitted to Redlich–Kister's type function in terms of mole fractions. Viscosities, speeds of sound and isentropic compressibilities of the binary mixtures have been correlated by means of several empirical and semi-empirical equations. The experimental data are analysed to discuss the nature and strength of intermolecular interactions in these mixtures.

Keywords: excess molar volume; viscosity; isentropic compressibility; 1-Butanol; chloroethanes; chloroethenes

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1. Introduction

This article is a continuation of our earlier work related to the study of thermodynamic and acoustical properties of binary mixtures [1–7]. In the earlier works we focused on the effect of $-CH_2$ group of alcohols on chloroalkanes and chloroalkenes [1], and the effect of aggregation between dimethyl sulfoxide and substituted alcohols [2]. The number of studies on thermodynamic properties of binary mixtures has increased in recent years due to industrial applications and the theoretical interest in studying the nature of molecular interaction and packing phenomena in binary mixtures. In this work, we measured densities, speeds of sound and viscosities of the binary mixtures of 1-butanol with chloroethanes, namely, 1,2-dichloroethane, 1,1,1-trichloroethane, 1,1,2,2,-tetrachloroethane, and chloroethenes, namely, trichloroethylene and tetrachloroethylene at 298.15 K and over the whole composition range. The data obtained are used to calculate excess molar volumes, viscosity deviations, deviation in isentropic compressibilities and excess Gibbs energies of activation of viscous flow of the binary mixtures. The results showed the negative values of viscosity deviations and excess Gibbs energies of activation and positive values of deviation in isentropic compressibilities and excess molar volumes for all the studied systems over the entire range of compositions.

The data of the binary systems were fitted to the Redlich–Kister [8] equation. The kinematic viscosity data for the binary mixtures were fitted to Nissan and Grunberg [9],

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Hind *et al*. [10], Katti and Chaudhri [11], McAllister [12] and Heric [13], and their parameters have been calculated. Theoretical models dealing with liquid state including, Schaaff collision factor theory (CFT) [14], Nomoto relation (NR) [15], Junjie relation (JR) [16] and Van Deal ideal mixing relation (IMR) [17] have been proposed for the prediction of the speeds of sound and isentropic compressibilities of the binary mixtures.

2. Experimental section

2.1. *Materials*

The purity of the chemicals is on the basis of mole%. All chemical substances were used without any further purification.

2.2. *Apparatus and procedure*

All mass measurements were performed on an electronic balance (AB 204-N Mettler) accurate to 0.1 mg. The uncertainty in the mole fractions was $\pm 1 \times 10^{-4}$. The densities of the pure compounds and their binary mixtures were measured with an Anton Paar DMA 4500 oscillating U-tube densimeter. The density measurement uncertainty was $\pm 1 \times 10^{-2}\,\mathrm{kg\,m^{-3}}$. The temperature in the cell was automatically regulated to ±0.01 K with a solid-state thermostat. The speed of sound in the pure compounds and their binary mixtures was measured with a multi-frequency ultrasonic interferometer supplied by Mittal Enterprise, New Delhi with an accuracy of $\pm 1\,\mathrm{m\,s^{-1}}$. In this work, 1 MHz frequency was employed. Viscosities at 298.15 K were measured with an Ubbleohde viscometer. The equation for viscosity is

$$\eta = \rho\nu = \rho\left(\frac{kt^2 - c}{t}\right), \tag{1}$$

where k and c are the viscometer constants and t, η and v are the efflux time, dynamic and kinematic viscosities, respectively. The dynamic viscosity was reproducible to within $\pm 2 \times 10^{-3}$ mPa s. The viscometer was suspended in a water bath maintained at ±0.01 K. Densities and refractive indices of the pure reagents were in good agreement with values found in the literature [18–20] (reported in Table 1).

Table 1. Source, purity grades, refractive index n_D, and density ρ, of pure component at 298.15 K.

Component	Source	Purity (mass%)	ρ (g cm^{-3})		n_D	
			Experiment	Literature	Experiment	Literature[a]
1-Butanol	Merck	99.5	0.80979	0.80970[a]	1.3991	1.3993
1,2-Dichloroethane	Merck	99.5	1.24564	1.24651[b]	1.4448	1.4443
1,1,1-Trichloroethane	Merck	98.0	1.3295	1.32963[b]	1.4379	1.4375
1,1,2,2-Tetrachloroethane	Fluka	99.0	1.5866	1.58683[b]	1.4940	1.4948
Trichloroethylene	Merck	99.5	1.45559	1.45572[c]	1.4773	1.4776
Tetrachloroethylene	Merck	99.0	1.61329	1.61309[c]	1.5053	1.5056

[a] From [18].
[b] From [19].
[c] From [20].

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3. Results and discussion

The density ρ, and V^E values for binary systems along with their densities ρ, at 298.15 K were measured and V^E against mole fraction of 1-butanol are graphically shown in Figure 1.

The excess molar volumes, V^E, were calculated from density data according to

$$V^{E} = \sum_{i=1}^{N} x_i M_i \left(\frac{1}{\rho} - \frac{1}{\rho_i} \right), \tag{2}$$

where x_i, M_i, and ρ_i are the mole fraction, molar mass, and density of the pure component i, respectively. ρ is the density of mixture, and N is the number of components. Figure 1 shows that the excess molar volumes are positive for all the studied systems over the whole composition range. The dependence of V^E on composition for the present mixtures may be explained as a balance between positive contributions hydrogen bond rupture and dispersive interactions between unlike molecules. In the present investigation, the interactions of butanol against chloroethanes and chloroethene molecules involve mainly dispersion force giving a positive contribution to V^E.

The viscosity deviations $\Delta\eta$ [21] and excess Gibbs energies of activation of viscous flow G^{*E} for binary mixtures can be calculated as:

$$\Delta\eta = \eta - \sum_{i=1}^{2} x_i \eta_i, \tag{3}$$

and

$$G^{*E} = RT\left(\ln\left(\frac{\eta V}{\eta_2 V_2} \right) - x_1 \ln\left(\frac{\eta_1 V_1}{\eta_2 V_2} \right) \right), \tag{4}$$

where V, V_1, and V_2 are the molar volumes of the binary mixture and pure component. The $\Delta\eta$ and G^{*E} values of all binary systems are calculated.

Figures 2 and 3 show the variation of $\Delta\eta$ and G^{*E} with the respect to mole fraction and volume fraction, respectively.

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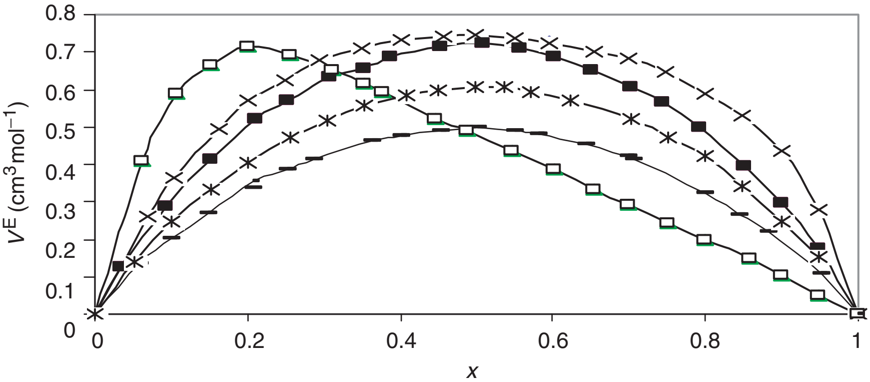

Figure 1. Exceess molar volumes for the binary mixtures of 1-butanol with 1,2-dichloroethane (×), 1,1,1-trichloroethane (■), 1,1,2,2-tetrachloroethane (□), trichloroethylene (–), and tetrachloroethylene (⋇) at 298.15 K. The solid curves were calculated from coefficients of Equation (5) given in Table 2.

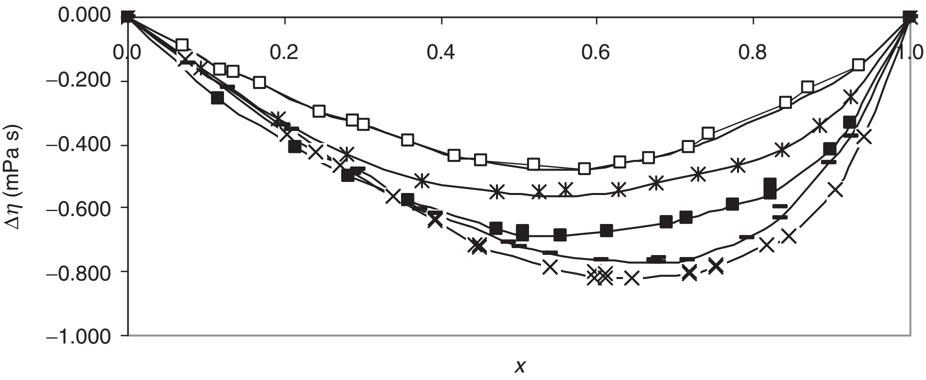

Figure 2. Viscosity deviation for the binary mixtures of 1-butanol with 1,2-dichloroethane (×), 1,1,1-trichloroethane (■), 1,1,2,2-tetrachloroethane (□), trichloroethylene (–) and tetrachloroethylene (⋇) at 298.15 K. The solid curves were calculated from coefficients of Equation (5) given in Table 2.

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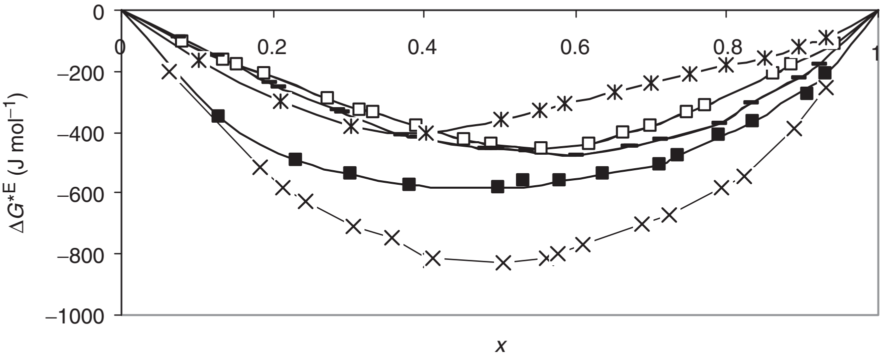

Figure 3. Excess Gibbs energies of activation for the binary mixtures of 1-butanol with 1,2-dichloroethane (×), 1,1,1-trichloroethane (■), 1,1,2,2-tetrachloroethane (□), trichloroethylene (–), and tetrachloroethylene (⋇) at 298.15 K. The solid curves were calculated from coefficients of Equation (5) given in Table 2.

Each set of results of V^{E}, $\Delta\eta$, and $\Delta\kappa_{\mathrm{s}}$ were fitted a Redlich–Kister polynomial [8], which for binary mixtures is

$$\Delta Y = x_1(1-x_1)\sum_{k=0}^{N} A_{\mathrm{k}}(2x_1-1)^{\mathrm{k}}, \tag{5}$$

where $\Delta Y \equiv V^{\mathrm{E}}$, $\Delta\eta$, $G^{*\mathrm{E}}$, or $\Delta\kappa_{\mathrm{s}}$. A_{k} is the polynomial coefficient obtained by a linear least-squares fitting procedure. In the case of $\Delta\kappa_{\mathrm{s}}$ the volume fraction, φ was considered instead of mole fraction. In each case, the optimum number of coefficients was ascertained from an examination of the variation of the standard deviation $\sigma(\Delta Y)$ with

$$\sigma(\Delta Y) = \left[\sum \frac{(\Delta Y_{\mathrm{exp}tl} - \Delta Y_{\mathrm{Calcd}})^2}{n-p}\right]^{1/2}, \tag{6}$$

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Table 2. Parameters of equation Redlich–Kister (5) and the standard deviations, σ in Equation (6) for 1-butanol with chloroethanes or chloroethenes at 298.15 K.

	A_0	A_1	A_2	A_3	A_4	A_5	σ
1-Butanol + 1,2-dichloroethene							
V^E ($cm^3 mol^{-1}$)	0.7479	1.5133	1.0971	0.6067	0.4113	0.1401	0.009
Δk_s ($T Pa^{-1}$)	−0.0183	79.775	−85.765	151.87	−276.39	130.47	0.3
$\Delta\eta$ (mPa s)	−0.0037	−1.0931	−6.8398	23.053	−28.587	13.459	0.002
G^{*E} ($J mol^{-1}$)	−9.5173	3022.4	−260.17	12,927	−17,552	7921.2	4.6
1-Butanol + 1,1,1-trichloroethane							
V^E ($cm^3 mol^{-1}$)	0.7165	1.4366	0.835	0.3134	0.3372	0.1411	0.009
Δk_s ($T Pa^{-1}$)	−0.0517	65.826	−69.858	184.11	−152.42	−28.871	0.6
$\Delta\eta$ (mPa s)	−0.0048	1.8354	−2.9389	16.183	−21.515	10.102	0.006
G^{*E} ($J mol^{-1}$)	−11.892	−3463.9	7251.7	−5500.5	−315.78	2034.9	5.9
1-Butanol + 1,1,2,2-tetrachloroethane							
V^E ($cm^3 mol^{-1}$)	0.4731	1.3961	1.9641	2.2812	1.9895	0.7568	0.008
Δk_s ($T Pa^{-1}$)	0.0453	153.27	72.567	−386.17	301.55	−141.1	0.2
$\Delta\eta$ (mPa s)	−0.0024	−1.3187	0.4831	−0.3182	2.5489	−1.4027	0.005
G^{*E} ($J mol^{-1}$)	−20.201	967.02	−306.16	−55.857	3543	−2201.5	8.2
1-Butanol + trichloroethylene							
V^E ($cm^3 mol^{-1}$)	0.5016	1.0092	0.3936	−0.1044	0.4341	0.4597	0.005
Δk_s ($T Pa^{-1}$)	0.0685	48	187.95	−507.9	405.22	−133.4	0.3
$\Delta\eta$ (mPa s)	−0.0039	1.5274	−2.4411	9.6471	−11.75	6.0725	0.009
G^{*E} ($J mol^{-1}$)	−8.741	−972.68	−2126	7255.4	−6858.3	2711.5	6.5
1-Butanol + tetrachloroethylene							
V^E ($cm^3 mol^{-1}$)	0.6083	1.2139	0.5567	−0.0178	0.3795	0.3826	0.004
Δk_s ($T Pa^{-1}$)	−0.0524	160.65	−101.19	69.563	−141.37	12.239	0.4
$\Delta\eta$ (mPa s)	−0.0014	−1.4717	−3.6968	17.845	−23.038	10.364	0.007
G^{*E} ($J mol^{-1}$)	9.1324	1205.1	−4403.5	21,318	−26,351	10,654	6.1

in which n is the number of results and p is the number of parameters retained in Equation (5). The adjustable parameters A_k, and standard deviations, σ are given in Table 2.

As can be observed in Figure 2, $\Delta\eta$ values are negative for all the binary mixtures over the whole mole fraction range. The $\Delta\eta$-containing mixtures change as follows:

1,2-Dichloroethane > Trichloroethylene > 1,1,1-Trichloroethane >
Tetrachloroethylene > 1,1,2,2-Tetrachloroethane

Isentropic compressibility k_s, was calculated using Newton–Laplace equation

$$k_s = \frac{1}{\rho u^2}. \tag{7}$$

Deviation in isentropic compressibilities $\Delta\kappa_s$, n-component were calculated using the following relation

$$\Delta k_s = k_s - \sum_i (k^*_{s,i}) x_i, \tag{8}$$

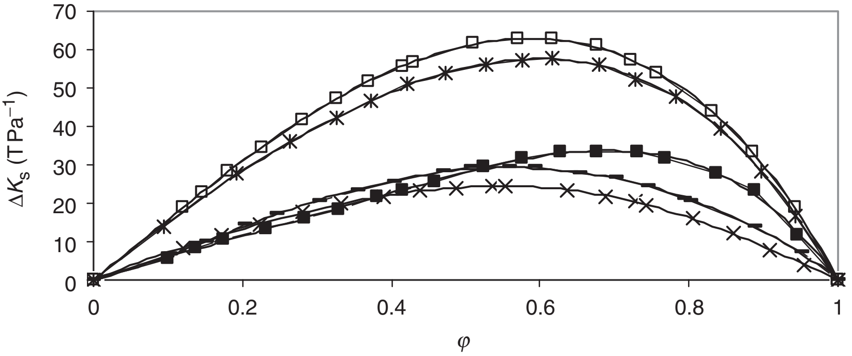

Figure 4. Variation of deviation in isentropic compressibility with volume fraction for the the mixtures of φ1-butanol + (1-φ)1,2-dichloroethane (×), 1,1,1-trichloroethane (■), 1,1,2,2-tetrachloroethane (□), trichloroethylene (–) and tetrachloroethylene (⁎) at 298.15 K. The solid curves were calculated from coefficients of Equation (5) given in Table 2.

where $k^*_{s,i}$ and k_s are the isentropic compressibility of pure components and mixtures, respectively. The densities ρ and speeds of sound u of the binary mixtures have been measured at 298.15 K. Corresponding isentropic compressibility, k_s, and deviation in isentropic compressibilities, Δk_s, are also calculated. The deviations in isentropic compressibility were plotted against volume fraction of φ,1-butanol in Figure 4.

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The speeds of sound were also calculated using the Schaaff collision factor theory (CFT) as following equations:

(i) Schaaff collision factor theory (CFT):

$$u = u_\infty \frac{\left(\sum_{i=1}^{n} x_i S_i\right)\left(\sum_{i=1}^{n} x_i B_i\right)}{V} = u_\infty S r_f, \tag{9}$$

where $u_\infty = 1600\,\mathrm{m\,s^{-1}}$, S = collision factor and $r_f = B/V$, space filling factor. B is the actual volume of the molecules per mole.

B_i can be evaluated as

$$B_i = \frac{4}{3}\pi r^3 N_A, \tag{10}$$

where r is the molecular radius of the pure component. Molecular radius is calculated as

$$r = \left[\frac{3b}{16\pi N_A}\right]^{1/3}, \tag{11}$$

where b is the van der Waal's constant.

(i) Nomoto relation (NR)

and

$$u = \left[\frac{\sum_{i=1}^{n} x_i R_i}{\sum_{i=1}^{n} x_i V_i}\right]^3, \tag{12}$$

$$R_i = V_i u_i^{1/3}, \tag{13}$$

Table 3. Standard deviations of predicted speeds of sound, and isentropic compressibility, by means of CFT, NR, JR and IMR of binary mixtures of 1-butanol with chloroethanes or chloroethenes at 298.15 K.

	CFT	NR	JR	IMR
1-Butanol + 1,2-dichloroethene				
$\sigma(u)$ (m s^{-1})	3.6	8.7	7.5	4.68
$\sigma(k_s)$ (T Pa^{-1})	7	13.2	11	8
1-Butanol + 1,1,1-trichloroethane				
$\sigma(u)$ (m s^{-1})	5.75	6.8	8.3	7.2
$\sigma(k_s)$ (T Pa^{-1})	8.3	11.6	12.8	10.2
1-Butanol + 1,1,2,2-tetrachloroethane				
$\sigma(u)$ (m s^{-1})	2.8	4.5	5.4	8.1
$\sigma(k_s)$ (T Pa^{-1})	4.3	6.2	8.5	10
1-Butanol + trichloroethylene				
$\sigma(u)$ (m s^{-1})	3.4	7.3	6	4.3
$\sigma(k_s)$ (T Pa^{-1})	9.4	14	12.9	11.25
1-Butanol + tetrachloroethylene				
$\sigma(u)$ (m s^{-1})	3.2	2.6	5.2	3.07
$\sigma(k_s)$ (T Pa^{-1})	7.6	8.2	9.3	7.01

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where R_i and u_i stand for Rao molar sound velocity and speed of sound of the pure component i, respectively.

(ii) Junjie relation (JR):

$$u = \frac{\sum_{i=1}^{n} x_i V}{\left(\sum_{i=1}^{n} x_i M_i\right)^{1/2}\left(\sum_{i=1}^{n} (x_i V_i/\rho_i u_i^2)\right)^{1/2}}. \tag{14}$$

(iii) Van Deal ideal mixing relation (IMR):

$$\left(\frac{1}{\sum_{i=1}^{n} x_i M_i}\right)\frac{1}{u^2} = \sum_{i=1}^{n} \frac{x_i}{M_i u_i^2}. \tag{15}$$

The parameter coefficients A_k, and standard deviations σ, for deviations in compressibility were also listed in Table 2.

The standard deviations for speeds of sound $\sigma(u)$, and isentropic compressibilities $\sigma(k_s)$ predicted by CFT, NR, JR and IMR are given in Table 3.

Figure 4 shows that the deviation in isentropic compressibilities, Δk_s, are the positive values for the entire range of composition, and increase as follows:

1,2-Dichloroethane > Trichloroethylene > 1,1,1-Trichloroethane >
Tetrachloroethylene > 1,1,2,2-Tetrachloroethane

These phenomena are results of differences in energies of interactions between molecules being in solution and packing effects. The order formation in the mixture (e.g. complex formation) leads to negative contribution for above-mentioned property.

Table 4. Adjustable parameters of Equations (16)–(20) and standard deviations of binary mixtures for viscosities of 1-butanol with chloroethanes or chloroethenes at 298.15 K.

Equation	System including 1-butanol +				
	1,2-Dichloroethane	1,1,1-Trichloroethane	1,1,2,2-Tetrachloroethane	Trichloroethylene	Tetrachloroethylene
Hind					
H_{12}	0.2319	0.4322	1.3678	0.2085	0.6973
σ	0.0038	0.0025	0.0046	0.0029	0.0023
Grunberg–Nissan					
G_{12}	−1.3951	−0.9847	−0.7013	−0.8068	−0.6234
σ	0.0010	0.0011	0.0023	0.0017	0.0024
Katti–Chaudri					
W	−5231.4	−4256.2	−4685.3	−3958.2	−3625.3
σ	0.0165	0.0143	0.0138	0.0163	0.0156
McAlister					
$\ln Z_{12}$	−11.2541	−11.5618	−11.3521	−11.6385	−11.7847
$\ln Z_{21}$	−11.4657	−11.2888	−11.2684	−11.9652	−11.7621
σ	0.00041	0.00036	0.00042	0.00017	0.00028
Heric					
α_{12}	0.3625	0.2195	−1.2365	−1.0012	−1.0865
α_{21}	0.023	−0.125	−0.423	−0.538	−0.625
σ	0.0017	0.0039	0.0043	0.0028	0.0051

The graphical presentation of the excess Gibbs energy of activation G^{*E}, shows the negative values for all the binary mixtures over the whole mole fraction range. According to Meyer *et al.* [22] negative values of G^{*E} correspond to the existence of solute–solute association.

To correlate experimental data of viscosities for binary systems, different empirical and semi-empirical relations have been used. The correlation equations are as follows:

(i) Nissan and Grunberg equation

$$\ln\eta = \sum_{i=1}^{n} x_i \ln\eta_i + \sum_{i=1}^{n}\sum_{j>i}^{n} x_i x_j A_{ij} + \sum_{i=1}^{n}\sum_{j>i}^{n}\sum_{k>j}^{n} x_i x_j x_k A_{ijk}. \tag{16}$$

(ii) Hind *et al.* equation

$$\eta = \sum_{i=1}^{n} x_i^2 \eta_i + 2\left(\sum_{i=1}^{n}\sum_{j>i}^{n} x_i x_j A_{ij} + \sum_{i=1}^{n}\sum_{j>i}^{n}\sum_{k>j}^{n} x_i x_j x_k A_{ijk}\right). \tag{17}$$

(iii) McAllister equation

$$\ln(\eta V) = \sum_{i=1}^{n} x_i^3 \ln(\eta_i V_i) + 3\sum_{i=1}^{n}\sum_{j\neq i}^{n} x_i^2 x_j \ln A_{ij} + 6\sum_{i=1}^{n}\sum_{j>i}^{n}\sum_{k>j}^{n} x_i x_j x_k \ln A_{ijk}. \tag{18}$$

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(iv) Katti and Chaudhri equation

$$\ln(\eta V) = \sum_{i=1}^{n} x_i \ln(\eta_i V_i) + \sum_{i=1}^{n}\sum_{j>i}^{n} x_i x_j A_{ij} + \sum_{i=1}^{n}\sum_{j>i}^{n}\sum_{k>j}^{n} x_i x_j x_k A_{ijk}. \tag{19}$$

(v) Heric equation

$$\ln(\eta V) = \sum_{i=1}^{n} x_i \ln(\eta_i V_i) + \sum_{i=1}^{n}\sum_{j>i}^{n} x_i x_j \left(\sum_{k=0}^{1} A_k \left(x_i - x_j\right)^k\right) + \sum_{i=1}^{n}\sum_{j>i}^{n}\sum_{k>j}^{n} x_i x_j x_k A_{ijk}. \tag{20}$$

Adjustable parameters of correlating equations and standard deviations are given in Table 4.

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