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Thermochimica Acta 427 (2005) 51–60

thermochimica acta

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Studies of viscosities of dilute solutions of alkylamine in non-electrolyte solvents. II. Haloalkanes and other polar solvents

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Received 1 May 2004; received in revised form 13 August 2004; accepted 13 August 2004
Available online 21 September 2004

Abstract

Viscosities η of dilute solutions of *n*-propylamine, *n*-butylamine, di-*n*-butylamine, and tri-*n*-propylamine in tetrachloromethane, *n*-propylamine, *n*-butylamine, di-*n*-propylamine, di-*n*-butylamine, tri-*n*-propylamine, and tri-*n*-butylamine in trichloromethane at 303.15 and 313.15 K and of triethylamine in 1,4-dioxane, ethyl ethanoate, butyl ethanoate, ethyl methyl ketone, chlorobenzene, and nitrobenzene have been measured at 303.15 K. The specific viscosities $(\eta - \eta_S^o)/\eta_S^o$ and viscosity deviations $\Delta\eta$ have been calculated. Based on Eyring's theory of absolute reaction rates, values of excess Gibbs energy of activation ΔG^{*E} of viscous flow have been determined. The results have been analyzed in terms of Herskovits and Kelly equation (J. Phys. Chem. 77 (1973) 381–388) and Nakagawa equation (J. Mol. Liq. 63 (1995) 303–316).

Keywords: Viscosities; Alkylamine; Non-electrolyte solvents; Herskovits and Kelly equation; Nakagawa equation

1. Introduction

As a part of our systematic studies on the thermodynamic and transport properties of dilute solutions of alkylamines in various solvents, in previous paper [1], measurements of viscosities of dilute solutions of *n*-propylamine (PA), *n*-butylamine (BA), di-*n*-propylamine (DPA), di-*n*-butylamine (DBA), tri-*n*-propylamine (TPA), and tri-*n*-butylamine (TBA) in cyclohexane and benzene and of triethylamine (TEA) in heptane, cyclohexane, benzene, toluene, ethylbenzene and *p*-xylene have been reported. As an extension of our studies in this paper we report viscosities η of PA, BA, DBA, and TPA in tetrachloromethane (CCl_4) and trichloromethane ($CHCl_3$), and DPA and TBA in $CHCl_3$ at 303.15 and 313.15 K and of TEA in 1,4-dioxane (DOX), ethyl ethanoate (EE), butyl ethanoate (BE), ethyl methyl ketone (EMK), chlorobenzene (CB), and nitrobenzene (NB), at 303.15 K. This study is expected to revel some interesting facts concerning the nature and different extent of interactions in amines. The results have also been analyzed in terms of Herskovits and Kelly equation [2] and Nakagawa equation [3].

2. Experimental

2.1. Method

The viscosities η were measured with a modified calibrated suspended level Ubbelohde viscometer [4]. The viscometer was designed so as to reduce surface tension effects to negligible values [5]. The densities ρ required to convert kinematic viscosities into dynamic viscosities η were measured by Anton Paar vibrating-tube digital densimeter (Model DMA 60/602) with thermostat bath controlled to ±0.01 K. The details of the experimental procedure have been described earlier [6,7]. Absorption curves were measured by means of a Schimadzu UV-160A double beam spectrophotometer, using 1 cm quartz cells.

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doi:10.1016/j.tca.2004.08.013

All the solutions were prepared by mixing known masses of pure liquids in air tight, narrow-mouth ground stoppered bottles taking due precautions to minimize the evaporation losses. Nine–ten solutions up to 0.1 mole fraction of each alkylamine in different solvents were prepared. The possible error in the mole fraction is estimated to be $<1 \times 10^{-4}$. The error in viscosity η was $<2 \times 10^{-3}$ mPa s.

2.2. Sources and purity of samples

PA (Merck, Schuchardt), BA (Fluka AG), DPA (Fluka AG), DBA (Fluka AG), TEA (Sisco, extra-pure), TPA (Fluka AG), and TBA (SRL, Bombay), of purity better than 99.0 mol.% were refluxed over Na metal and distilled twice fractionally before use. CCl_4 (BDH, AR), $CHCl_3$ (BDH, AR), DOX (Merck, AR), EE (Galaxo, AR), BE (Galaxo, AR), EMK (Galaxo, AR), CB (Merck, AR) and NB (Merck, AR) of purity better than 99.5 mol.% were used after further purification and drying by standard procedures [8]. The measured values of density and viscosities were compared with literature values in Table 1.

Table 1
Comparison of experimental densities and viscosity of pure liquids at 303.15 K with the literature data

Solvents	Temp. (K)	Density ($g\,cm^{-3}$)		Viscosity (mPa s)	
		Obs.	Lit.	Obs.	Lit.
PA	303.15	0.70610	0.70615[a]	0.353	0.350[b]
	313.15	0.69604	0.6960[a]	0.313	0.311[c]
BA	303.15	0.72865	0.72715[d]	0.444	0.456[b] 0.458[e]
	313.15	0.71917	0.7182[f] 0.7192[g]	0.393	0.3831[f] 0.398[e]
DPA	303.15	0.73121	0.73019[h]	0.512	0.483[i] 0.490[c]
	313.15	0.72615	0.7216[a]	0.423	0.425[i] 0.435[c]
DBA	303.15	0.75228	0.75248[e]	0.758	0.734[j] 0.75009[j]
	313.15	0.74344	0.74284[j] 0.7431[d]	0.665	0.64[j] 0.727[i]
TEA	303.15	0.71844	0.71845[b] 0.71875[a]	0.330	0.3295[b] 0.3298[k] 0.328[i]
TPA	303.15	0.74915	0.7484[d]	0.598	0.577[c]
	313.15	0.74041	0.7404[d]	0.538	0.505[c]
TBA	303.15	0.77021	0.7701[l]	1.167	1.1663[k] 0.7709[a]
	313.15	0.76311 0.99[j]	0.7612[j]	0.970	0.989[k] 0.7634[d]
CCl_4	303.15	1.57478	1.57480[b]	0.852	0.845[b]
	313.15	1.55502	1.5555[m]	0.760	0.736[b]
$CHCl_3$	303.15	1.47064	1.47060[b]	0.520	0.514[b]
	313.15	1.44843	1.45203[b]	0.471	0.467[b]
DOX	303.15	1.0223	1.0223[b]	1.086	1.087[b]
EE	303.15	0.88842	0.88843[b]	0.399	0.400[b]
BE	303.15	0.87128	0.87129[b]	0.644	0.644[b]
EMK	303.15	0.79969	0.7997[b]	0.365	0.366[b]
CB	303.15	1.09549	1.09550[b]	0.718	0.7184[b]
NB	303.15	1.19340	1.19341[b]	1.617	1.619[b]

[a] [9]; [b] [8]; [c] [10]; [d] [11]; [e] [12]; [f] [13]; [g] [14]; [h] [15]; [i] [16]; [j] [17]; [k] [18]; [l] [19]; [m] [20].

3. Theoretical

3.1. Herskovits and Kelly equation

In general, for interpreting viscosity data the following relation:

$$\frac{\eta}{\eta_S^o} = 1 + Bm + Dm^2 \tag{1}$$

has commonly been used [21], where η/η_S^o is the relative viscosity, B a coefficient related to the size and shape of solute molecule and to solvation effects, whereas D the coefficient includes solute–solute interactions and those solute–solvent interactions which arise with higher concentrations and are not accounted for by the Bm term.

Herskovits and Kelly [2] substituted molality in terms of volume faction ϕ_s as $m = 1000\phi_s/M_s\bar{v}_s\rho_S$ in Eq. (1) and obtained Eq. (2):

$$\frac{\eta}{\eta_S^o} = 1 + \frac{1000B}{M_s\bar{v}_s\rho_S}\phi_s + \frac{(1\times 10^6)D}{(M_s\bar{v}_s\rho_S)^2}{\phi_s}^2 \tag{2}$$

where $\bar{v}_s$ is the partial specific volume. The coefficients B and D of equation can be evaluated from the intercept and slope of $(\eta/\eta_S^o - 1)/m$ versus m plots.

3.2. Nakagawa equation

Recently, Matsubayashi and Nakahara [22] have extended the conformal solution theory (ECS) [23] to the dynamic problem through the first-order perturbation theory, theoretically derived the formula about concentration dependence of dynamical properties, and shown the validity of their ECS theory for binary regular solution containing benzene. According to the ECS theory, a dynamic property η for a binary mixture is written as follows:

$$\eta = x_s\eta_s^o + (1 - x_s)\eta_S^o + (1 - x_s)x_s\eta_{int} \tag{3}$$

where η_s^o and η_S^o are viscosities of solute and solvent components and η_{int} is the interaction term for regular solution.

3.2.1. Derivation of B coefficients from η_{int} through ECS theory

For non-electrolyte solutions of higher concentrations, Jones–Dole equation [24,25] can be rewritten as:

$$\frac{\eta}{\eta_S^o} = 1 + B'c + D'c^2 \tag{4}$$

where c is the concentrations (molarity; mol dm^{-3}) and B' and D' are adjustable parameters. Differentiating Eq. (4) by molarity c at constant temperature T and pressure P,

$$\left[\frac{\partial(\eta/\eta_{\mathrm{S}}^{\mathrm{o}})}{\partial c}\right]_{T,P} = B' + 2D'c \tag{5}$$

Form Eq. (5), the B' coefficient is the first partial differential coefficient by molarity at $c = 0$, i.e.

$$B' = \lim_{c\to 0}\left[\frac{\partial(\eta/\eta_{\mathrm{S}}^{\mathrm{o}})}{\partial c}\right]_{T,P,c=0} \tag{6}$$

Eq. (3) can be rearranged to

$$\frac{\eta}{\eta_{\mathrm{S}}^{\mathrm{o}}} = x_{\mathrm{s}}\left(\frac{\eta_{\mathrm{s}}^{\mathrm{o}}}{\eta_{\mathrm{S}}^{\mathrm{o}}}\right) + (1 - x_{\mathrm{s}}) + (1 - x_{\mathrm{s}})x_{\mathrm{s}}\frac{\eta_{\mathrm{int}}}{\eta_{\mathrm{S}}^{\mathrm{o}}} \tag{7}$$

Since, the interaction term η_{int} is independent to the concentration for regular solution, we get the following equation by differentiating Eq. (7) by x_{s} at constant T and P.

$$\left[\frac{\partial(\eta/\eta_{\mathrm{S}}^{\mathrm{o}})}{\partial x_{\mathrm{s}}}\right]_{T,P} = \left(\frac{\eta_{\mathrm{s}}^{\mathrm{o}}}{\eta_{\mathrm{S}}^{\mathrm{o}}}\right) - 1 + (1 - 2x_{\mathrm{s}})\left(\frac{\eta_{\mathrm{int}}}{\eta_{\mathrm{S}}^{\mathrm{o}}}\right) \tag{8}$$

The mole fraction x_{s} and concentration c of solute in molarity are related with following relation:

$$c = \frac{1000x_{\mathrm{s}}\rho}{x_{\mathrm{s}}M_{\mathrm{s}} + (1 - x_{\mathrm{s}})M_{\mathrm{S}}} \tag{9}$$

The density ρ of solution can be expressed as power series of x_{s} [22], that is,

$$\rho = \sum_{i=0}^{n} A_i x_{\mathrm{s}}^{i} \tag{10}$$

where A_i is fitting coefficient, in particular ($A_0 \equiv \rho_{\mathrm{S}}$) means the neat solvent density. Substituting Eq. (10) in Eq. (9), and differentiating it by x_{s} at constant T and P, one obtains:

$$\left[\frac{\partial c}{\partial x_{\mathrm{s}}}\right]_{T,P} = \frac{1000\left[\sum\limits_{i=1}^{n} A_i x_{\mathrm{s}}^{i+1}(M_{\mathrm{s}} - M_{\mathrm{S}}) + \sum\limits_{i=0}^{n}(i + 1)A_i x_{\mathrm{s}}^{i} M_{\mathrm{S}}\right]}{[x_{\mathrm{s}}M_{\mathrm{s}} + (1 - x_{\mathrm{s}})M_{\mathrm{S}}]^2} \tag{11}$$

Combination of Eqs. (8) and (11), and comparison with Eq. (6), gives

$$B' = \lim_{c\to 0}\left[\frac{\partial(\eta/\eta_{\mathrm{S}}^{\mathrm{o}})}{\partial c}\right] = \lim_{c\to 0}\frac{[\partial(\eta/\eta_{\mathrm{S}}^{\mathrm{o}})/\partial x_{\mathrm{s}}]}{[\partial c/\partial x_{\mathrm{s}}]} \tag{12}$$

$$B' = \frac{M_{\mathrm{S}}[(\eta_{\mathrm{s}}^{\mathrm{o}}/\eta_{\mathrm{S}}^{\mathrm{o}}) - 1 + (\eta_{\mathrm{int}}/\eta_{\mathrm{S}}^{\mathrm{o}})]}{1000\rho_{\mathrm{S}}} \tag{13}$$

3.2.2. Division of B' coefficient

For an ideal solution, namely, $\eta_{\mathrm{int}} = 0$, Eq. (13) is written as

$$B'_{\mathrm{id}} = \frac{M_{\mathrm{S}}[(\eta_{\mathrm{s}}^{\mathrm{o}}/\eta_{\mathrm{S}}^{\mathrm{o}}) - 1]}{1000\rho_{\mathrm{S}}} \tag{14}$$

where B'_{id} is the contribution of ideal mixture for the B' coefficient, which is based on the difference between the viscosity of solute and solvent. If η_{s} is larger than η_{S}, the B'_{id} coefficient is positive and vice versa. This finding implies that the B' coefficient is not characteristic for solute–solvent interaction.

It is proposed that $B'_{\mathrm{int}} = B' - B'_{\mathrm{id}}$, namely,

$$B'_{\mathrm{int}} = \frac{M_{\mathrm{S}}\eta_{\mathrm{in}}}{1000\rho_{\mathrm{S}}\eta_{\mathrm{S}}} = K\eta_{\mathrm{int}} \tag{15}$$

should be the interaction parameter instead of B', where $K = M_{\mathrm{S}}/1000\rho_{\mathrm{S}}\eta_{\mathrm{S}} = 1/1000\upsilon_{\mathrm{S}}\eta_{\mathrm{S}}$ is the constant which is independent of the solution composition.

4. Results

The experimental viscosities η, specific viscosities $(\eta - \eta_{\mathrm{S}}^{\mathrm{o}})/\eta_{\mathrm{S}}^{\mathrm{o}}$, viscosity deviations $\Delta\eta$ and excess Gibbs energy of activation ΔG^{*E} of viscous flow of the dilute solutions of PA, BA, DBA, and TPA in CCl_4 and $CHCl_3$, and DPA and TBA in $CHCl_3$ at 303.15 and 313.15 K and of TEA in DOX, EE, BE, EMK, CB, and NB at 303.15 K are presented in Table 2. TEA solutions in CCl_4 and $CHCl_3$ were not investigated because of reaction between the components. The solutions were found to be opaque at the conclusion of the experiment.

The experimental values of η are expressed by:

$$\eta = \sum_{i=1}^{m} A_i x_{\mathrm{s}}^{i-1} \tag{16}$$

The coefficients A_i obtained from a least-squares fit with equal weights assigned to each point, are listed in Table 3 together with the standard deviations σ.

Since components used in the solution formation are both liquids and each one of them has dynamic viscosity. It would be appropriate to take into account viscosity of both the components in calculating viscosity deviations from a linear dependence on mole fraction. The viscosity deviations $\Delta\eta$ have been evaluated as:

$$\Delta\eta = \eta - (x_{\mathrm{s}}\eta_{\mathrm{s}}^{\mathrm{o}} + x_{\mathrm{S}}\eta_{\mathrm{S}}^{\mathrm{o}}) \tag{17}$$

On the basis of the theory of absolute reaction rates [26], the excess Gibbs energy of activation ΔG^{*E} of viscous flow has been calculated from:

$$\frac{\Delta G^{*E}}{RT} = \left\{\ln(\eta V/\eta_{\mathrm{S}}^{\mathrm{o}}V_{\mathrm{S}}^{\mathrm{o}}) - x_{\mathrm{s}}\ \ln(\eta_{\mathrm{s}}^{\mathrm{o}}V_{\mathrm{s}}^{\mathrm{o}}/\eta_{\mathrm{S}}^{\mathrm{o}}V_{\mathrm{S}}^{\mathrm{o}})\right\} \tag{18}$$

where η and V are the viscosity and molar volume of the solution; and $\eta_{\mathrm{s}}^{\mathrm{o}}$ and $\eta_{\mathrm{S}}^{\mathrm{o}}$ the viscosities and $V_{\mathrm{s}}^{\mathrm{o}}$ and $V_{\mathrm{S}}^{\mathrm{o}}$ are the molar volumes of the pure solute and solvent, respectively. The values of $\Delta\eta$ and ΔG^{*E} have been fitted to the polynomial of the form:

$$\psi = \sum_{i=1}^{m} A_i x_{\mathrm{s}}^{i} \tag{19}$$

Table 2
Properties of amine solutions in different solvents

x_s	m (mol kg^{-1})	303.15 K		313.15 K	
		ρ (g cm^{-3})	η (mPa s)	ρ (g cm^{-3})	η (mPa s)
CCl_4 + PA					
0.0071	0.0468	1.56984	0.859	1.55016	0.761
0.0184	0.1222	1.56186	0.866	1.54230	0.763
0.0285	0.1907	1.55457	0.873	1.53507	0.764
0.0386	0.2610	1.54737	0.876	1.52792	0.765
0.0479	0.3275	1.54078	0.880	1.52137	0.768
0.0574	0.3962	1.53392	0.885	1.51452	0.770
0.0661	0.4598	1.52748	0.885	1.50811	0.772
0.0748	0.5258	1.52124	0.888	1.50194	0.774
0.0841	0.5967	1.51430	0.892	1.49514	0.775
CCl_4 + BA					
0.0099	0.0647	1.56674	0.855	1.54713	0.763
0.0183	0.1212	1.55988	0.858	1.54041	0.765
0.0287	0.1924	1.55136	0.861	1.53201	0.768
0.0386	0.2614	1.54306	0.862	1.52390	0.770
0.0477	0.3253	1.53531	0.862	1.51618	0.773
0.0581	0.4009	1.52657	0.864	1.50758	0.776
0.0675	0.4706	1.51867	0.866	1.49984	0.780
0.0760	0.5347	1.51152	0.869	1.49272	0.784
0.0856	0.6084	1.50349	0.871	1.48469	0.786
CCl_4 + DBA					
0.0105	0.0691	1.56001	0.852	1.54058	0.761
0.0201	0.1333	1.54659	0.851	1.52731	0.760
0.0298	0.1999	1.53323	0.850	1.51426	0.761
0.0396	0.2682	1.51986	0.850	1.50114	0.760
0.0490	0.3349	1.50712	0.849	1.48859	0.759
0.0583	0.4023	1.49477	0.847	1.47640	0.757
0.0696	0.4863	1.48000	0.846	1.46190	0.755
0.0789	0.5570	1.46795	0.845	1.45012	0.754
0.0890	0.6356	1.45515	0.845	1.43755	0.752
CCl_4 + TPA					
0.0103	0.0680	1.55808	0.848	1.53854	0.754
0.0203	0.1344	1.54201	0.840	1.52271	0.747
0.0304	0.2039	1.52635	0.829	1.50724	0.739
0.0398	0.2692	1.51185	0.819	1.49294	0.733
0.0497	0.3398	1.49693	0.811	1.47828	0.726
0.0606	0.4192	1.48086	0.802	1.46246	0.718
0.0695	0.4855	1.46802	0.795	1.44976	0.712
0.0786	0.5545	1.45503	0.788	1.43701	0.706
0.0893	0.6378	1.44017	0.780	1.42230	0.699
$CHCl_3$ + PA					
0.0100	0.0848	1.46310	0.523	1.44112	0.474
0.0207	0.1767	1.45477	0.526	1.43307	0.478
0.0296	0.2557	1.44798	0.529	1.42656	0.481
0.0426	0.3724	1.43789	0.533	1.41680	0.485
0.0491	0.4329	1.43295	0.536	1.41195	0.488
0.0595	0.5299	1.42484	0.539	1.40397	0.491
0.0689	0.6201	1.41767	0.542	1.39696	0.495
0.0796	0.7241	1.40935	0.544	1.38875	0.496
0.0960	0.8901	1.39686	0.548	1.37641	0.500
$CHCl_3$ + BA					
0.0091	0.0771	1.46278	0.528	1.44074	0.478
0.0196	0.1671	1.45353	0.536	1.43189	0.485
0.0289	0.2497	1.44549	0.544	1.42381	0.493
0.0409	0.3571	1.43501	0.550	1.41372	0.499
0.0483	0.4248	1.42852	0.556	1.40741	0.505
0.0578	0.5135	1.42029	0.562	1.39924	0.511
0.0676	0.6072	1.41170	0.566	1.39072	0.517
0.0775	0.7041	1.40316	0.569	1.38230	0.521

Table 2 (*Continued*)

x_s	m (mol kg^{-1})	303.15 K		313.15 K	
		ρ (g cm^{-3})	η (mPa s)	ρ (g cm^{-3})	η (mPa s)
0.0848	0.7766	1.39676	0.573	1.37601	0.525
$CHCl_3$ + DPA					
0.0105	0.0888	1.45785	0.525	1.43595	0.473
0.0217	0.1854	1.44432	0.531	1.42268	0.476
0.0311	0.2688	1.43319	0.535	1.41178	0.480
0.0411	0.3590	1.42135	0.539	1.40029	0.483
0.0510	0.4503	1.40985	0.544	1.38904	0.486
0.0608	0.5418	1.39852	0.548	1.37803	0.488
0.0708	0.6378	1.38722	0.552	1.36694	0.492
0.0808	0.7358	1.37600	0.558	1.35601	0.494
0.0914	0.8426	1.36432	0.561	1.34460	0.497
$CHCl_3$ + DBA					
0.0103	0.0871	1.45546	0.530	1.43369	0.480
0.0197	0.1682	1.44184	0.540	1.42036	0.489
0.0294	0.2538	1.42811	0.552	1.40690	0.493
0.0381	0.3319	1.41597	0.560	1.39499	0.502
0.0482	0.4238	1.40196	0.572	1.38138	0.510
0.0593	0.5277	1.38711	0.584	1.36685	0.516
0.0684	0.6149	1.37509	0.599	1.35519	0.528
0.0788	0.7164	1.36157	0.612	1.34211	0.535
0.0876	0.8038	1.35035	0.624	1.33113	0.542
$CHCl_3$ + TPA					
0.0106	0.0898	1.45358	0.531	1.43177	0.476
0.0206	0.1759	1.43745	0.541	1.41602	0.480
0.0320	0.2767	1.41970	0.546	1.39873	0.485
0.0396	0.3455	1.40799	0.554	1.38754	0.489
0.0509	0.4491	1.39116	0.563	1.37093	0.493
0.0596	0.5313	1.37863	0.568	1.35879	0.501
0.0693	0.6240	1.36478	0.579	1.34521	0.504
0.0787	0.7160	1.35178	0.590	1.33244	0.511
0.0883	0.8111	1.33863	0.598	1.31939	0.516
$CHCl_3$ + TBA					
0.0102	0.0864	1.45010	0.533	1.42849	0.479
0.0205	0.1752	1.43019	0.545	1.40915	0.488
0.0308	0.2663	1.41092	0.557	1.39040	0.496
0.0404	0.3528	1.39358	0.569	1.37358	0.504
0.0505	0.4460	1.37608	0.583	1.35650	0.513
0.0598	0.5328	1.36044	0.593	1.34135	0.522
0.0695	0.6252	1.34477	0.606	1.32607	0.529
0.0799	0.7277	1.32842	0.615	1.31005	0.538
0.0856	0.7843	1.31972	0.621	1.30156	0.545
DOX + TEA					
0.0138	0.1590	1.01532	1.059		
0.0232	0.2697	1.01065	1.042		
0.0349	0.4098	1.00486	1.018		
0.0453	0.5388	0.99987	0.998		
0.0551	0.6616	0.99517	0.980		
0.0691	0.8419	0.98857	0.955		
0.0803	0.9909	0.98341	0.932		
0.0909	1.1350	0.97857	0.914		
0.1006	1.2695	0.97422	0.899		
0.1128	1.4433	0.96878	0.877		
EE + TEA					
0.0127	0.1465	0.88547	0.395		
0.0205	0.2373	0.88369	0.389		
0.0323	0.3793	0.88100	0.386		
0.0407	0.4809	0.87910	0.382		
0.0522	0.6256	0.87653	0.379		
0.0608	0.7349	0.87462	0.376		

Table 2 (*Continued*)

x_s	m (mol kg^{-1})	303.15 K		313.15 K	
		ρ (g cm^{-3})	η (mPa s)	ρ (g cm^{-3})	η (mPa s)
0.0702	0.8575	0.87241	0.374		
0.0783	0.9644	0.87058	0.371		
0.0900	1.1229	0.86791	0.369		
0.0996	1.2551	0.86605	0.367		
BE + TEA					
0.0065	0.0564	0.87030	0.641		
0.0144	0.1257	0.86910	0.638		
0.0226	0.1988	0.86785	0.631		
0.0293	0.2600	0.86683	0.628		
0.0378	0.3385	0.86553	0.623		
0.0454	0.4090	0.86442	0.618		
0.0509	0.4620	0.86354	0.613		
0.0596	0.5456	0.86222	0.607		
0.0668	0.6161	0.86113	0.603		
0.0733	0.6808	0.86014	0.599		
EMK + TEA					
0.0110	0.1543	0.79896	0.362		
0.0217	0.3080	0.79822	0.360		
0.0315	0.4511	0.79760	0.357		
0.0413	0.5972	0.79687	0.355		
0.0508	0.7423	0.79632	0.353		
0.0597	0.8808	0.79550	0.351		
0.0697	1.0394	0.79480	0.347		
0.0801	1.2071	0.79425	0.344		
0.0915	1.3975	0.79337	0.341		
0.1022	1.5787	0.79279	0.338		
CB + TEA					
0.0118	0.1056	1.08987	0.709		
0.0193	0.1753	1.08626	0.702		
0.0299	0.2735	1.08132	0.694		
0.0415	0.3849	1.07587	0.685		
0.0493	0.4606	1.07221	0.677		
0.0595	0.5621	1.06748	0.672		
0.0710	0.6787	1.06215	0.664		
0.0792	0.7642	1.05837	0.658		
0.0893	0.8707	1.05374	0.652		
0.0994	0.9804	1.04913	0.645		
NB + TEA					
0.0113	0.0925	1.18681	1.590		
0.0201	0.1668	1.18170	1.569		
0.0288	0.2408	1.17668	1.547		
0.0404	0.3417	1.16997	1.521		
0.0506	0.4330	1.16425	1.499		
0.0601	0.5190	1.15870	1.480		
0.0688	0.6006	1.15374	1.466		
0.0819	0.7245	1.14633	1.440		
0.0910	0.8129	1.14121	1.422		
0.1005	0.9076	1.13648	1.407		

Table 3
Coefficients A_i of Eq. (16) along with standard deviation σ for viscosity

Solvents	Temp. (K)	A_1	A_2	A_3	σ (mPa s)
CCl_4 + PA	303.15	0.8538	0.7146	−3.2836	0.0012
	313.15	0.7599	0.1384	0.6130	0.0008
CCl_4 + BA	303.15	0.8530	0.2308	−0.3427	0.0012
	313.15	0.7604	0.2239	0.9601	0.0006
CCl_4 + DBA	303.15	0.8530	−0.0910	−0.0577	0.0005
	313.15	0.7603	0.0492	−1.6504	0.0006
CCl_4 + TPA	303.15	0.8555	−0.9064	0.5855	0.0017
	313.15	0.7607	−0.7038	0.1095	0.0005
$CHCl_3$ + PA	303.15	0.5201	0.3310	−0.3648	0.0007
	313.15	0.4704	0.3966	−0.8784	0.0007
$CHCl_3$ + BA	303.15	0.5206	0.8751	−3.0675	0.0008
	313.15	0.4708	0.7913	−1.7726	0.0007
$CHCl_3$ + DPA	303.15	0.5207	0.4584	−0.1352	0.0006
	313.15	0.4703	0.3075	−0.1474	0.0006
$CHCl_3$ + DBA	303.15	0.5209	0.8947	3.6547	0.0029
	313.15	0.4719	0.7463	0.6497	0.0016
$CHCl_3$ + TPA	303.15	0.5227	0.7092	1.6036	0.0018
	313.15	0.4713	0.3876	1.3827	0.0010
$CHCl_3$ + TBA	303.15	0.5201	1.2664	−0.8897	0.0013
	313.15	0.4710	0.7960	0.6845	0.0007
DOX + TEA	303.15	1.0866	−2.0120	1.3481	0.0010
EE + TEA	303.15	0.3994	−0.4740	1.5033	0.0008
BE + TEA	303.15	0.6446	−0.5373	−1.2938	0.0009
EMK + TEA	303.15	0.3648	−0.2158	−0.4705	0.0005
CB + TEA	303.15	0.7184	−0.8686	1.3484	0.0008
NB + TEA	303.15	1.6180	−2.5715	4.7215	0.0012

Table 4
Coefficients A_i of Eq. (19) along with standard deviations σ for viscosity deviations

Solvents	Temp. (K)	A_1	A_2	A_3	σ (mPa s)
CCl_4 + PA	303.15	1.3986	−9.2275	47.8919	0.0010
	313.15	0.5888	0.1793	5.0161	0.0009
CCl_4 + BA	303.15	0.7787	−4.0734	26.3122	0.0009
	313.15	0.6738	−1.8080	22.6461	0.0008
CCl_4 + DBA	303.15	−0.0092	0.4923	−4.7820	0.0007
	313.15	0.2043	−3.5185	14.4262	0.0005
CCl_4 + TPA	303.15	−0.1079	−16.4941	132.4334	0.0022
	313.15	−0.3321	−4.6960	37.6617	0.0007
$CHCl_3$ + PA	303.15	0.3358	4.2107	−32.1976	0.0004
	313.15	0.4507	2.0186	−20.2973	0.0006
$CHCl_3$ + BA	303.15	0.8538	0.1418	−25.8610	0.0009
	313.15	0.8469	−1.1770	−4.2054	0.0008
$CHCl_3$ + DPA	303.15	0.3943	2.3276	−19.6170	0.0008
	313.15	0.2028	4.5779	−35.8800	0.0008
$CHCl_3$ + DBA	303.15	0.6163	5.2276	−13.5330	0.0031
	313.15	0.7460	−5.5158	48.4351	0.0018
$CHCl_3$ + TPA	303.15	0.9708	−8.6788	78.4221	0.0017
	313.15	0.4026	−1.3911	22.4403	0.0012
$CHCl_3$ + TBA	303.15	0.5258	2.7975	−26.4750	0.0010
	313.15	0.2853	1.1815	−4.4883	0.0008
DOX + TEA	303.15	−1.1779	−0.4064	10.2183	0.0010
EE + TEA	303.15	−0.3004	−1.8139	24.5390	0.0012
BE + TEA	303.15	−0.0952	−5.7506	39.7793	0.0008
EMK + TEA	303.15	−0.2326	1.0245	−10.4242	0.0006
CB + TEA	303.15	−0.3882	−1.4565	20.3306	0.0011
NB + TEA	303.15	−1.2355	2.9597	13.7023	0.0016

The coefficients A_i of Eq. (19), obtained by the method of least squares with all points weighted equally, and the standard deviations σ are given in Tables 4 and 5.

5. Discussion

We have determined specific viscosities $(\eta - \eta_S^o)/\eta_S^o$, viscosity deviations $\Delta\eta$ and excess Gibbs energies of activation

Table 5
Coefficients A_i of Eq. (19) along with standard deviations σ for ΔG^{*E}

Solvent	Temp. (K)	A_1	A_2	σ (J mol^{-1})
CCl_4 + PA	303.15	4591.5	−14051.3	5.0
	313.15	2687.4	2863.5	2.8
CCl_4 + BA	303.15	2504.8	−4070.7	4.1
	313.15	2537.5	1452.2	2.6
CCl_4 + DBA	303.15	480.2	−633.0	1.5
	313.15	1098.6	−7804.8	2.5
CCl_4 + TPA	303.15	−115.8	−12470.8	12.2
	313.15	−422.7	−5946.5	4.2
$CHCl_3$ + PA	303.15	2060.5	3950.5	7.2
	313.15	2798.8	−280.4	6.3
$CHCl_3$ + BA	303.15	4311.0	−12271.1	4.7
	313.15	4682.7	−10075.3	3.6
$CHCl_3$ + DPA	303.15	2477.8	108.9	3.4
	313.15	1916.5	3872.1	6.6
$CHCl_3$ + DBA	303.15	4184.0	11902.6	12.1
	313.15	4696.1	−8722.1	10.7
$CHCl_3$ + TPA	303.15	5170.7	−8755.7	13.0
	313.15	3214.1	1566.0	5.6
$CHCl_3$ + TBA	303.15	5889.9	−8639.9	6.5
	313.15	4710.1	−4117.3	3.6
DOX + TEA	303.15	−1167.5	−2637.0	3.0
EE + TEA	303.15	−2062.9	4632.1	7.2
BE + TEA	303.15	−237.4	−9237.6	3.5
EMK + TEA	303.15	−1322.6	−1821.1	4.7
CB + TEA	303.15	−883.4	1333.6	3.8
NB + TEA	303.15	−6.3	4911.4	2.2

ΔG^{*E} of flow at 303.15 K as well as at 313.15 K for some solutions. The dependence of $(\eta - \eta_S^0)/\eta_S^0$ on m, $\Delta\eta$ and ΔG^{*E} on x_s is shown only at 303.15 K, since for other temperatures the curves are similar. The values of $(\eta - \eta_S^0)/\eta_S^0$ are positive and their magnitude increases with increase in amine mole fractions, except for DBA and TPA solutions in tetrachloromethane and of TEA in 1,4-dioxane, ethyl ethanoate, butyl ethanoate, ethyl methyl ketone, chlorobenzene, and nitrobenzene. In general, the plots of $(\eta - \eta_S^0)/\eta_S^0$ versus m are almost linear (Figs. 1–3) for amine solutions in different solvents except some curvature is observed for PA, BA and DPA in $CHCl_3$. Values of slope of these curves depend on the size and shape of solute molecules and to the solvation effects. The relative values of $\Delta\eta$ and ΔG^{*E} have been analyzed in terms of solute–solute and solute–solvent interactions.

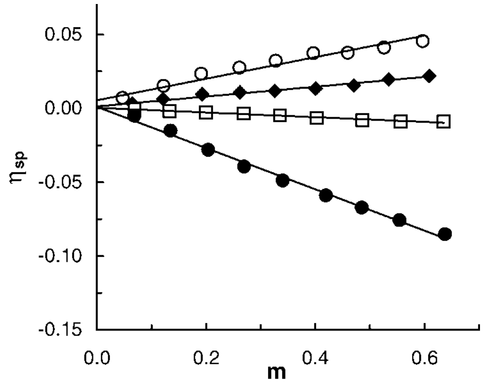

Fig. 1. Specific viscosities of: (○) PA, (♦) BA, (□) DBA, and (●) TPA in CCl_4 at 303.15 K.

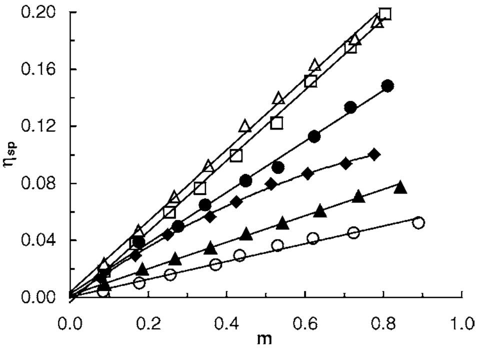

Fig. 2. Specific viscosities of: (○) PA, (♦) BA, (▲) DPA, (□) DBA, (●) TPA and (△) TBA in $CHCl_3$ at 303.15 K.

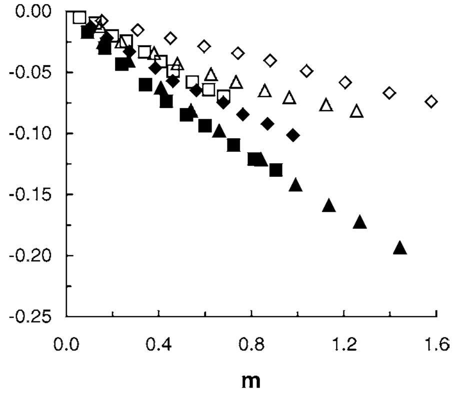

Fig. 3. Specific viscosities of TEA in: (▲) DOX, (△) EE, (□) BE, (◇) EMK, and (♦) CB and (■) NB at 303.15 K.

5.1. Alkylamine in tetrachloromethane

Figs. 4 and 5 show that the values of $\Delta\eta$ and ΔG^{*E} are positive for PA and BA and negative for TPA in CCl_4. For DBA in CCl_4, they are very small positive or negative

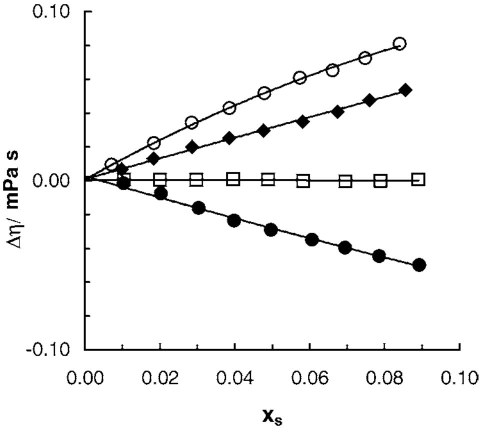

Fig. 4. Viscosity deviations of: (○) PA, (♦) BA, (□) DBA, and (●) TPA in CCl_4 at 303.15 K.

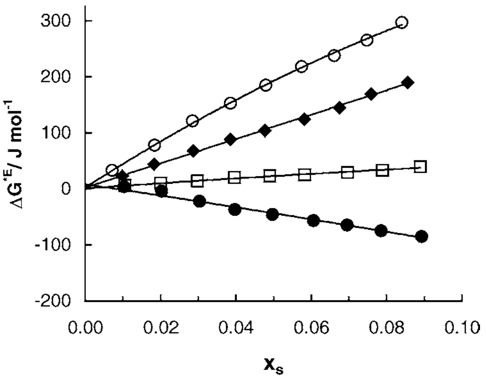

Fig. 5. Excess Gibbs energy of activation of viscous flow for: (○) PA, (♦) BA, (□) DBA, and (●) TPA in CCl_4 at 303.15 K.

within almost experimental uncertainty. The values of $\Delta\eta$ and ΔG^{*E} increase for primary (PA and BA) while decrease for secondary (DBA) and tertiary amine (TPA) solutions with the increase of amine concentration. The positive values of $\Delta\eta$ and ΔG^{*E} definitely indicate N···Cl specific interactions [27] which are stronger when primary and secondary amines are involved than those for tertiary amines. There are thermodynamic evidences for complex formation between tetrachloromethane and butylamine or dibutylamine. Complex formation between amine and tetrachloromethane is well supported by very large negative excess enthalpies and large negative excess molar volumes [28–30]. The values of H^{E}_{min} and V^{E}_{min} for BA + CCl_4 are $-4616\,J\,mol^{-1}$ at $x_s = 0.7762$ [28] and $-0.52\,cm^3\,mol^{-1}$ at $x_s = 0.55$ [29]. The values of V^{E}_{min} for DBA + CCl_4 is $-0.723\,cm^3\,mol^{-1}$ at $x_s = 0.70$ [30]. Larger negative H^E values for BA + CCl_4 compared to TEA + CCl_4 ($\sim -700\,J\,mol^{-1}$ [31]) suggest stronger interaction in BA + CCl_4. Stevenson and Coppinger [32] and Foster [33] have reported that the reaction of triethylamine, di-*n*-propylamine and tri-*n*-propylamine with tetrachloromethane is photochemical, forming corresponding alkyl amine hydrochloride and chloroform. In order to see whether specific interaction is through charge transfer, as observed between CCl_4 and TEA from UV-spectroscopic results by Stevenson and Coppinger [32], we have also performed similar experiments of tetrachloromethane and amine solutions. The spectra of 1% (v/v) solution of CCl_4, amines and CCl_4 + amine mixtures in cyclohexane are given in Fig. 6(a) and (b). As may be seen that curves of spectra of amines in tetrachloromethane show red shift from the cyclohexane solution. This spectral behaviour of amines in tetrachloromethane solutions indicates charge transfer complex between tetrachloromethane and presently investigated amines. Thus, like in TEA + CCl_4 [32], tetrachloromethane forms charge transfer complexes with other amines too.

5.2. Alkylamine in trichloromethane

Figs. 7 and 8 show that values of $\Delta\eta$ and ΔG^{*E} are large and positive for all the six amine solutions in trichloromethane. According to Fort and Moore [27] such results are indicative of specific interaction leading to complex formations. Specific interactions have also been reported from excess molar volumes for dibutylamine + trichloromethane [30]. Specific interactions have also been predicted from large negative H^E ($-3818\,J\,mol^{-1}$) and V^E ($\sim -1.9\,cm^3\,mol^{-1}$) values for $(C_2H_5)_3N$ + $CHCl_3$ mixtures [34]. These specific interactions can be of N···H–C and N···Cl–C types. UV-spectral study of 1% (v/v) solution of $CHCl_3$, amines

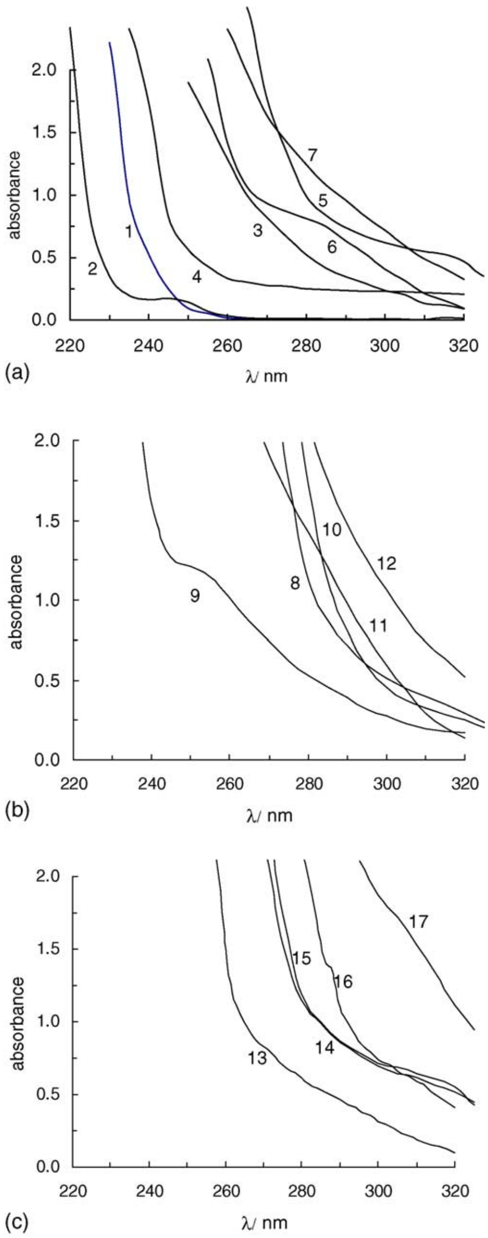

Fig. 6. UV-absorption spectra of CCl_4, $CHCl_3$, amines, CCl_4 + amine, $CHCl_3$ + amine solutions in cyclohexane. (a) Curves 1–7 are for CCl_4, $CHCl_3$, PA, BA, DBA, TPA and TBA, respectively, in cyclohexane solution; (b) Curves 8–12 are for CCl_4 + PA, CCl_4 + BA, CCl_4 + DBA, CCl_4 + TPA and CCl_4 + TBA, respectively, in cyclohexane solution; (c) Curves 13–17 are for $CHCl_3$ + PA, $CHCl_3$ + BA, $CHCl_3$ + DBA, $CHCl_3$ + TPA and $CHCl_3$ + TBA, respectively, in cyclohexane solution.

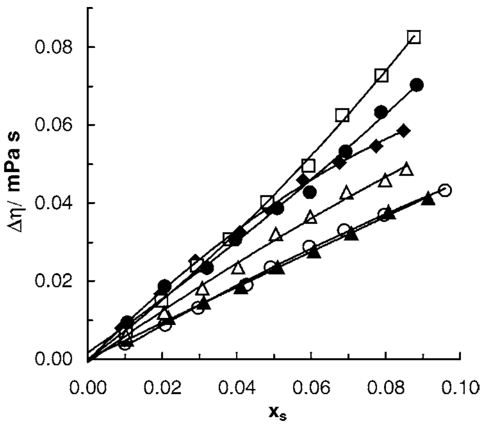

Fig. 7. Viscosity deviations of: (○) PA, (♦) BA, (▲) DPA, (□) DBA, (●) TPA and (△) TBA in $CHCl_3$ at 303.15 K.

and $CHCl_3$ + amines mixture in cyclohexane (Fig. 6(a) and (c)) shows expected red shift for amines in trichloromethane solution, confirming the charge transfer complex between trichloromethane and amines. Both the types of specific interactions N···H–C and N···Cl–C have been identified for mixtures of triethylamine and chloroform, H-bond interaction from ^{1}H-NMR [35] and charge transfer interaction by UV [32]. Thus, it may be inferred that both N···H–C and N···Cl–C types of interactions play a decisive role in such solutions.

5.3. Triethylamine in other polar solvents

As far as 1,4-dioxane solutions are concerned, the value of $\Delta\eta$ and ΔG^{*E} are large and negative (Figs. 9 and 10) and are comparable to cyclohexane solutions [1]. Thus, like cyclohexane molecules, the 1,4-dioxane molecules acts as an order breaker for TEA and shows negative deviations in viscosity behaviour. The value of $\Delta\eta$ and ΔG^{*E} are negative and their magnitude increases with increase in TEA mole fraction in DOX, EE, BE, EMK, CB, and NB (Figs. 9 and 10). Since the latter solvents are highly polar possessing considerable dipole moment. Therefore, it is expected to observe strong dipole–dipole interactions between TEA and these solvents [36,37]. According to Fort and Moore [27] predominance of specific interactions is expected to result positive deviations in the viscosity. The present observations are opposite to the expected behaviour. It is known that the strength of the specific interactions is not the only factor influencing the viscosity deviation in solutions, but the molecular size and shape of the components are also equally important. Similar were the observations in many amine–alcohol solutions [38,39].

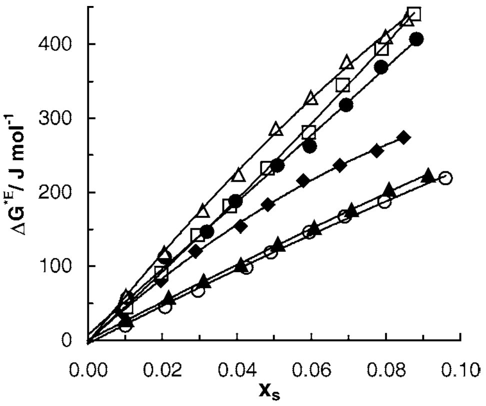

Fig. 8. Excess Gibbs energy of activation of viscous flow for: (○) PA, (♦) BA, (▲) DPA, (□) DBA, (●) TPA and (△) TBA in $CHCl_3$ at 303.15 K.

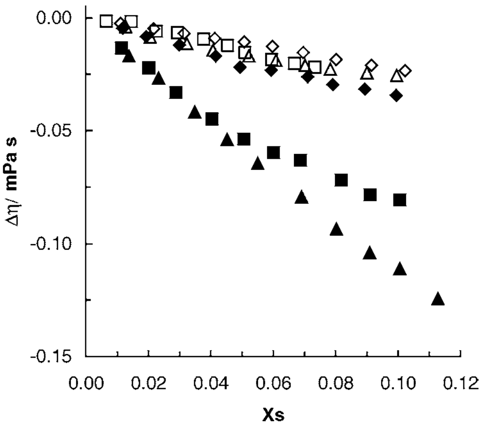

Fig. 9. Viscosity deviations of TEA in: (▲) DOX, (△) EE, (□) BE, (◇) EMK, and (♦) CB and (■) NB at 303.15 K.

5.4. On Herskovits and Kelly equation

Table 6 gives that the values of coefficient B and D of Eq. (2), viscosity increment $\nu = 1000\phi_S/M_S\bar{v}_S\rho_S$ and the values of $10^6 D/(M_S\bar{v}_S\rho_S)^2$ in presently investigated amine solutions. The values of partial specific volume of amines $\bar{v}_S$ reported in VI column have been derived from the measurements of densities of dilute solutions at number of concentrations of amines [40,41]. The values of B coefficient are in the range from −0.126 to −0.145 for alkyl amines in CCl_4, 0.054 to 0.270 in $CHCl_3$, and −0.048 to −0.193 in DOX,

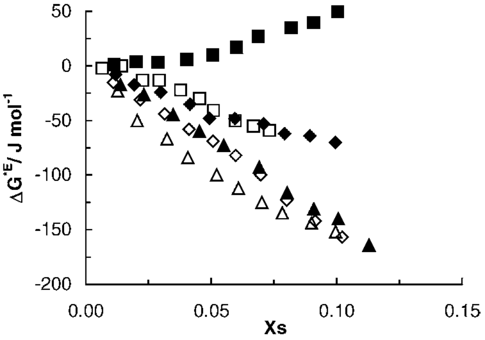

Fig. 10. Excess Gibbs energy of activation of viscous flow for TEA in: (▲) DOX, (△) EE, (□) BE, (◇) EMK, (♦) CB and (■) NB at 303.15 K.

Table 6
B and D coefficients and standard deviations σ estimated from Eq. (2) and the values of partial specific volume $\bar{v}_s$, viscosity increment $\upsilon = 1000\phi_s/M_s\bar{v}_s\rho_S$ and $10^6 D/(M_s\bar{v}_s\rho_S)^2$

Systems	Temp. (K)	B	D	σ	$\bar{v}_s$	υ	$10^6 D/(M_s\bar{v}_s\rho_S)^2$
CCl_4 + PA	303.15	0.145	−0.128	0.002	1.36	1.14	−8.01
	313.15	0.027	0.014	0.001	1.38	0.21	0.86
CCl_4 + BA	303.15	0.058	−0.044	0.002	1.32	0.38	−1.90
	313.15	0.055	0.001	0.001	1.33	0.36	0.03
CCl_4 + DBA	303.15	−0.017	0.002	0.001	1.32	−0.06	0.03
	313.15	0.019	−0.062	0.001	1.33	0.07	−0.87
CCl_4 + TPA	303.15	−0.111	−0.060	0.005	1.35	−0.36	−0.65
	313.15	−0.126	−0.007	0.002	1.36	−0.42	−0.08
$CHCl_3$ + PA	303.15	0.054	0.014	0.003	1.39	0.44	0.92
	313.15	0.084	−0.011	0.002	1.40	0.70	−0.79
$CHCl_3$ + BA	303.15	0.186	−0.072	0.002	1.34	1.29	−3.46
	313.15	0.195	−0.060	0.002	1.35	1.36	−2.95
$CHCl_3$ + DPA	303.15	0.097	−0.003	0.002	1.35	0.48	−0.08
	313.15	0.058	0.016	0.003	1.37	0.29	0.40
$CHCl_3$ + DBA	303.15	0.204	0.065	0.006	1.32	0.81	1.04
	313.15	0.215	−0.042	0.005	1.33	0.87	−0.67
$CHCl_3$ + TPA	303.15	0.206	−0.043	0.006	1.31	0.75	−0.56
	313.15	0.109	0.009	0.002	1.33	0.40	0.12
$CHCl_3$ + TBA	303.15	0.270	−0.024	0.003	1.29	0.77	−0.20
	313.15	0.201	−0.004	0.001	1.30	0.58	−0.03
DOX + TEA	303.15	−0.158	0.017	0.001	1.40	−1.09	0.81
EE + TEA	303.15	−0.093	0.022	0.003	1.38	−0.75	1.40
BE + TEA	303.15	−0.083	−0.037	0.002	1.38	−0.68	−2.53
EMK + TEA	303.15	−0.048	0.001	0.002	1.33	−0.30	0.05
CB + TEA	303.15	−0.128	0.024	0.001	1.36	−0.85	1.07
NB + TEA	303.15	−0.193	0.055	0.001	1.34	−1.19	2.11

EE, BE, EMK, CB, and NB. The values of viscosity increment $\upsilon = 1000B/M_s\bar{v}_s\rho_S$ are both positive and negative for amine solutions in CCl_4, positive for $CHCl_3$ while negative for amine solutions in DOX, EE, BE, EMK, CB, and NB. On the basis of Einstein model of sphere in a continuum [42], the viscosity increment υ should be equal to 2.5. The values of viscosity increment υ are not only far different than 2.5 but also have negative values. The deviation of B coefficient and related viscosity increments from the Einstein [42] and Guth and Simha [43] values may be attributed to the nature of solvent in terms of structure forming and structure breaking, and the deviation of solute particle from sphericity. Lower values of υ were also found for homologous series of non-electrolytes in non-polar solvents [1,21].

5.5. *On Nakagawa equation*

We have estimated the B', $B'_{\rm id}$ and $B'_{\rm int}$ coefficients from the $\eta_{\rm int}$ values (Table 7) using Eqs. (13)–(15). The values of B' coefficient for alkylamine are from −0.092 to 0.074 in CCl_4, from 0.047 to 0.190 in $CHCl_3$ and −0.065 to −0.158 in other solvents. Since the B' coefficient also contains the contributions to the difference of viscosities for solute and solvent components and, therefore, it is inappropriate to discuss the solute–solvent interaction in terms of B'. Solute–solvent interactions can be considered by the term $B'_{\rm int}$ defined by Eq. (15).

Table 7
Regular terms of viscosities $\eta_{\rm int}$, viscosity B', $B'_{\rm id}$, $B'_{\rm int}$ parameters

System	Temp. (K)	$\eta_{\rm int}$	B'	$B'_{\rm id}$	$B'_{\rm int}$
CCl_4 + PA	303.15	1.150	0.074	−0.057	0.132
	313.15	0.642	0.025	−0.058	0.084
CCl_4 + BA	303.15	0.693	0.033	−0.047	0.079
	313.15	0.687	0.042	−0.048	0.089
CCl_4 + DBA	303.15	0.001	−0.011	−0.011	0.000
	313.15	0.077	−0.002	−0.012	0.010
CCl_4 +TPA	303.15	−0.545	−0.092	−0.029	−0.062
	313.15	−0.473	−0.090	−0.029	−0.061
$CHCl_3$ + PA	303.15	0.468	0.047	−0.026	0.073
	313.15	0.512	0.062	−0.028	0.090
$CHCl_3$ + BA	303.15	0.824	0.116	−0.012	0.128
	313.15	0.817	0.129	−0.014	0.143
$CHCl_3$ + DPA	303.15	0.474	0.072	−0.001	0.074
	313.15	0.337	0.051	−0.008	0.059
$CHCl_3$ + DBA	303.15	0.877	0.174	0.037	0.137
	313.15	0.655	0.149	0.034	0.115
$CHCl_3$ + TPA	303.15	0.823	0.140	0.012	0.128
	313.15	0.425	0.086	0.012	0.074
$CHCl_3$ + TBA	303.15	0.614	0.190	0.094	0.096
	313.15	0.348	0.148	0.087	0.061
DOX + TEA	303.15	−1.230	−0.158	−0.060	−0.098
EE + TEA	303.15	−0.324	−0.098	−0.017	−0.080
BE + TEA	303.15	−0.256	−0.118	−0.065	−0.053
EMK + TEA	303.15	−0.230	−0.065	−0.009	−0.057
CB + TEA	303.15	−0.413	−0.115	−0.056	−0.059
NB + TEA	303.15	−1.075	−0.151	−0.082	−0.069

The values of B'_{int} are in general positive for all amine solutions in CCl_4 and $CHCl_3$, except for TPA in CCl_4, which clearly indicate specific interactions between amine and haloalkane. On comparison of B'_{int} for TEA solutions in cyclohexane [1] and TEA solutions in DOX, EE, BE, EMK, CB, and NB, it is observed that negative value of B'_{int} is reduced, which may be attributed to the presence of specific interactions between solute–solvent. The specific interactions may be considered as dipole–dipole interactions. The negative values of B'_{int} further reveals that in these solutions dispersion interactions are dominating over the solute–solvent specific interactions. The interpretation based on the relative magnitude of B'_{int} derived from the ECS theory is consistent with the partial molar volume and the heat of solution.

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