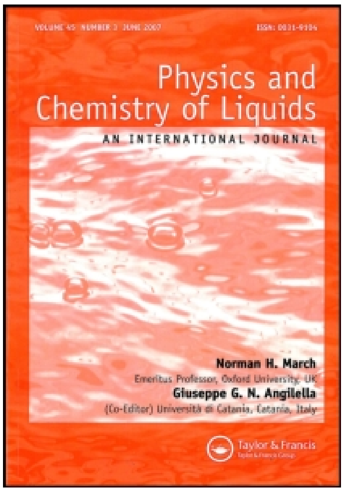

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VISCOSITY BEHAVIOUR OF 2-BUTOXYETHANOL -N-HEXANE AND -N-HEPTANE MIXTURES AT 303.15 AND 313.15 K

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Excess viscosities and free energy of activation of flow have been reported for the binary liquid mixtures of 2-butoxyethanol with n-hexane and n-heptane at 303.15 and 313.15 K. The $\Delta \ln \eta^E$ and ΔG^E are positive. Bloomfield and Dewan approach based on Flory theory is used to calculate the various thermodynamic contributions to the mixtures viscosities. Entropy contributions are found to contribute to the observed experimental excess viscosities. The calculations indicate that the disorder type interactions are predominant in the present systems studied. The mixture viscosities are also found to be reasonably expressed by Grunberg-Nissan and Mc Allister relations.

KEY WORDS: Viscosities, free energy of flow of activation, entropy contributions, disorder interactions.

1. INTRODUCTION

Alkoxyethanols especially the ethoxy- and butoxyethanols are widely used industrial solvents. The simultaneous presence of proton donating -OH group and proton accepting etheric linkage in the same molecule make interesting interactions at molecular level in the binary mixtures containing alkoxyethanols as one of the components. Inspite of the greater attention, the alkoxyethanols received as solvents, there exist relatively few studies involving measurements of thermo- dynamic properties on binary mixtures of alkoxyethanols with other polar and nonpolar solvents.

Casanova *et al.*, have reported the excess enthalpies[1,2], excess molar volumes and heat capacities[3,4] of n-alkoxyethanols-organic solvent mixtures. An attempt was also made to give a quantitative interpretation of excess molar volumes and isentropic compressibilities of 2-ethoxyethanol – n-heptane[5], -glycol[6] and -water[7] mixtures interms of specific molecular interactions. Mac Neil *et al.*[8] have also indicated the presence of weak hydrogen bonding interactions between 2-butoxyethanol and several anilines from density and viscosity measurements. Koga *et al.*[9–12] have dealt in detail the aqueous solutions of 2-butoxyethanols through measurements of various thermodynamic properties such as vapour pressure, partial molar volumes and heat capacities. The authors have suggested the presence of three regions in each of which the mixing scheme is qualitatively different from that in the other regions. Similar observation was also made by Fioretto *et al.*[13,14] from dielectric relaxation measurements.

We have recently reported the excess dielectric functions of 2-butoxyethanol–n-hexane, and n-heptane mixtures[15]. In continuation of our systematic studies of the thermodynamic properties of binary mixtures with alkoxyethanols as one of the components, this paper describes the measurements on viscosities and densities of 2-butoxyethanol–n-hexane and -n-heptane mixtures both at 303.15 and 313.15 K. The excess viscosity functions such as $\Delta \ln \eta^E$ and free energy of activation of flow, ΔG^E are derived from the mixture viscosity and density data. The mixture viscosities were also derived from Flory theory based on Bloomfield–Dewan approach to ascertain the dominant factor that contributes to the mixture viscosity. The mixture viscosities were also derived from Grunberg–Nissan and Mc Allister's approaches.

2. EXPERIMENTAL SECTION

Materials

2-Butoxyethanol was of Fluka puriss grade (with > 99% purity on mole basis) and used as such without any further treatment. n-Hexane and n-heptane were also of SD AnalaR reagent grade and were used with out any further purification procedures. The purity of the chemicals was checked by measuring their densities, viscosities and refractive indices and comparing them with literature values. Such a comparison is presented in Table 1.

The binary solutions were prepared in glass vials by mass on a single pan Mettler balance with a mole fraction accuracy of ± 0.0001.

The densities of the pure components and their mixtures were determined by a bicapilary pyknometer. The details of the calibration of the pyknometer was described elsewhere[16]. The measured densities were accurate to ±0.0001 units.

Table 1 Densities (ρ), Viscosities (η) and Refractive Indices (n_D) of pure liquids.

	298.15 K		*303.15 K*		*313.15 K*	
	This work	*Lit.*	*This work*	*Lit.*	*This work*	*Lit.*
			n-Hexane			
$\rho, g.\mathrm{cm}^{-3}$	0.6547	0.6548(26) 0.6545(27)	0.6502	0.6502(26) 0.6506(27)	0.6411	0.6414(34) 0.6413(27)
η, cP	0.292	0.292(34) 0.294(35)	0.264	----	0.237	----
n_D	1.3723	1.3722(28)	1.3695	1.3698(26) 1.3699(29)	1.3675	1.3642(29)
			n-Heptane			
$\rho, g.\mathrm{cm}^{-3}$	0.6794	0.6794(30)	0.6756	0.6754(26)	0.6660	0.6668(31)
η, cP	0.392	0.390(28)	0.376	----	0.335	----
n_D	1.3850	1.3851(28)	1.3830	1.3827(26)	1.3800	----
			2-Butoxyethanol			
$\rho, g.\mathrm{cm}^{-3}$	0.8963	0.8962(8) 0.8960(32)	0.8924	0.8920(8) 0.8929(33)	0.8834	0.8835(33) 0.8836(8)
η, cP	2.784	2.786(28)	2.401	2.403(8)	1.868	1.867(8)
n_D	----	----	1.4300	----	1.4109	----

The viscosities, η were measured by an Ubbelhode suspended type viscometer. The viscometer was calibrated with double distilled benzene and triple distilled water at the measuring temperatures to evaluate the viscometer constants A and B, which were further used to calculate the absolute viscosities from the measured flow times t by the following relation

$$\eta = \rho(A - B/t) \tag{1}$$

where ρ = density in g.cm^{-3}. The measured viscosities were accurate upto ±0.003 cP.

THEORY

Flory Theory

The mixture viscosities can also be theoretically predicted by combining the absolute reaction rate theory of Eyring and co-workers and free volumes theory. Bloomfield and Dewan[17] had described the following relation,

$$\ln\eta = x_1 \ln\eta_1 + x_2 \ln\eta_2 - \frac{\Delta H_m}{RT} + \frac{\Delta S^R}{R} + \left(\frac{1}{\tilde{v}-1} - \frac{x_1}{\tilde{v}_1 - 1} - \frac{x_2}{\tilde{v}_2 - 1}\right) \tag{2}$$

where ΔH_m is the enthalpy of mixing for the mole of the solution, ΔS^R is the residual entropy per mole, R is the gas constant T is the absolute temperature in K. $\tilde{v}$, $\tilde{v}_1$ and $\tilde{v}_2$ are the reduced volumes of the mixture and pure components respectively. The Eq. 2 can also be written alternately as

$$\Delta \ln\eta^E = \ln\eta_H + \ln\eta_S + \ln\eta_V \tag{3}$$

where

$$\ln\eta_H = -\frac{\Delta H_m}{RT},$$

$$\ln\eta_S = \frac{\Delta S^R}{R}$$

and

$$\ln\eta_V = \frac{1}{\tilde{v}-1} - \frac{x_1}{\tilde{v}_1 - 1} - \frac{x_2}{\tilde{v}_2 - 1}$$

The individual enthalpy and entropy contributions to the mixture viscosity can be obtained by the relations given by Flory and co-workers[18,19] as per Eq. 4 and Eq. 5

$$\frac{\Delta H_m}{RT} = \frac{x_1 C_1}{\tilde{T}_1}\left(\frac{1}{\tilde{v}} - \frac{1}{\tilde{v}_1}\right) + \frac{x_2 C_2}{\tilde{T}_2}\left(\frac{1}{\tilde{v}} - \frac{1}{\tilde{v}_2}\right) + \frac{x_1 C_1 \theta \chi_{12}}{\tilde{v}\,\tilde{T}_1 P_1^*} \tag{4}$$

$$\frac{\Delta S^R}{R} = -3\left[x_1 C_1 \ln\frac{\tilde{v}_1^{1/3} - 1}{\tilde{v}^{1/3} - 1} + x_2 C_2 \ln\frac{\tilde{v}_2^{1/3} - 1}{\tilde{v}^{1/3} - 1}\right] \tag{5}$$

The various terms that are given in Eqs. 4 and 5 are same as described in detail by Abe and Flory[19] in their original papers. The reduced volumes of the mixtures and the pure components are calculated with the use of experimental densities, thermal expansion coefficients, α and the molar volumes V using following relations

$$\tilde{v} = \frac{V}{x_1 V_1^* + x_2 V_2^*}, \quad \tilde{v}_i = \frac{V_i}{V_i^*} \tag{6}$$

The characteristic parameters viz. V^*, temperature T^* and pressure P^* have the same significance as described by Abe and Flory. The parameter C_i for a component is defined as

$$C_i = \frac{P_i^* V_i^*}{R\,T_i^*} \tag{7}$$

The parameter χ_{12} that was employed for the evaluation of enthalpy contribution was estimated from the fit of experimental excess volume data at equimolar concentrations.

The characteristic parameters of the pure liquids together with other physicochemical data are given in Table 2.

Grunberg-Nissan approach

The viscosity of the mixture is also expressed by Eq. 8 by Grunberg and Nissan[20,21]

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x(1-x)\,d_{12} \tag{8}$$

d_{12} in Eq. 8 is an adjustible parameter and is taken as a measure of strength of the interaction between the components in the mixture.

Mc Allister approach

The composition dependence of the absolute viscosity for the binary mixtures is also expressed by following Eq. 9 based on three-body interaction model proposed by Mc Allister[22].

$$\begin{aligned} \ln \eta = {} & x_1^3 \ln \eta_1 + 3x_1^2 x_2 \ln \upsilon_{12} + 3x_1 x_2^2 \ln \upsilon_{21} + x_2^3 \ln \eta_2 + x_2^3 \ln (M_2/M_1) \\ & + 3x_1 x_2^2 \ln \{(1+2M_2)/M_1)/3\} + 3x_1^2 x_2 \ln \{(2+M_2)/M_1/3\} \\ & - \ln (x_1 + (x_2 M_2/M_1) \end{aligned} \tag{9}$$

Table 2 Physico-Chemical Data and Equation of State Parameters for Pure Liquids at 303.15 K.

Liquid	$\alpha \times 10^3 K^{-1}$	$\tilde{v}$	V^* *cm*^{3} *mol*$^{-1}$	P^* *J. cm*$^{-3}$	$\tilde{T}$	*Vcm*^{3} *mol*$^{-1}$
Hexane	1.404	1.3292	99.71	419.5	0.06808	132.45
Heptane	1.266	1.3038	113.74	426.8	0.06491	148.39
Butoxy-ethanol	0.964	1.2436	106.48	475.9	0.05636	132.43

The terms η, x and M are the viscosity, mole fraction and molecular weight of the component 1 and 2 respectively. The terms υ_{ij} are the adjustible parameters and are determined from the fits of experimental viscosity data by least square procedure.

RESULTS AND DISCUSSION

The experimental densities and viscosities for the binary mixtures at 303.15 and 313.15 K are presented in Table 3. The logarithmic excess viscosities, $\Delta \ln\eta^E$ and excess free energy of flow, ΔG^E as evaluated from the following relations

$$\Delta \ln\eta^E = \ln\eta_{12} - (x_1 \ln\eta_1 + x_2 \ln\eta_{12}) \tag{10}$$

$$\Delta G^E = RT(\ln\eta_{12} V_{12} - x_1 \ln\eta_1 V_1 - x_2 \ln\eta_2 V_2) \tag{11}$$

where η and V are the viscosities and molar volumes of the pure components 1 or 2 and mixture 12, are also included in Table 3.

Table 3 Densities (ρ), Viscosities (η), Excess viscosities ($\Delta \ln\eta^E$) and Free energy of flow of activation (ΔG^E) of the 2-butoxyethanol (1)-alkane mixtures at 303.15 and 313.15 K.

	$T = 303.15\ K$				$T = 313.15\ K$			
x_1	$\rho\ g.cm^{-3}$	$\eta\ cP$	$\Delta \ln\eta^E cP$	$\Delta G^E J.mol^{-1}$	$\rho\ g.cm^{-3}$	$\eta\ cP$	$\Delta \ln\eta^E cP$	$\Delta G^E J.mol^{-1}$
				2-Butoxyethanol-n-hexane				
0.0327	0.6576	0.292	0.028	74.10	0.6484	0.258	0.018	48.95
0.1014	0.6738	0.354	0.069	178.10	0.6645	0.310	0.060	154.25
0.1974	0.6965	0.456	0.110	284.55	0.6842	0.396	0.105	273.46
0.2974	0.7196	0.587	0.143	368.48	0.7109	0.506	0.144	371.76
0.3950	0.7437	0.741	0.160	410.32	0.7344	0.634	0.168	433.00
0.4968	0.7683	0.930	0.162	416.62	0.7589	0.787	0.175	448.13
0.5957	0.7924	1.148	0.155	369.21	0.7830	0.956	0.165	423.17
0.7323	0.8259	1.507	0.125	320.59	0.8166	1.230	0.135	345.55
0.8316	0.8505	1.793	0.080	204.15	0.8412	1.458	0.100	253.80
0.8992	0.8673	2.025	0.052	133.83	0.8587	1.634	0.074	148.00
0.9658	0.8840	2.271	0.020	50.41	0.8748	1.790	0.028	71.33
				2-Butoxyethanol-n-heptane				
0.0365	0.6812	0.407	0.012	35.29	0.6725	0.364	0.019	55.72
0.1077	0.6947	0.484	0.053	179.70	0.6859	0.426	0.055	149.96
0.2126	0.7152	0.626	0.116	303.20	0.7063	0.534	0.100	272.06
0.3160	0.7361	0.799	0.168	436.53	0.7272	0.661	0.136	367.36
0.4170	0.7572	0.987	0.192	497.52	0.7482	0.797	0.150	591.00
0.5114	0.7775	1.177	0.190	530.29	0.7685	0.941	0.151	444.16
0.6155	0.8006	1.411	0.181	469.04	0.7915	1.116	0.146	390.79
0.7469	0.8308	1.741	0.148	381.84	0.8216	1.359	0.117	313.20
0.8450	0.8542	2.011	0.110	283.80	0.8448	1.550	0.080	214.27
0.9057	0.8690	2.162	0.070	180.08	0.8597	1.654	0.040	109.35
0.9663	0.8839	2.306	0.022	58.47	0.8748	1.784	0.012	33.85

The excess viscosity functions are fitted through an empirical relation

$$Y^E = x_2(1 - x_2)\sum a_i(2x_2 - 1)^i \tag{12}$$

where Y^E = either $\Delta \ln \eta^E$ or ΔG^E respectively. The constants a_i are evaluated by a regression analysis. The values of the constants together with the standard deviations, σ are given in Table 4.

In Fig. 1, are shown the experimental $\Delta \ln \eta^E$ at 303.15 and 313.15 K together with the calculated values through Eq. 12 and the constants from the Table 4 as a function of 2-butoxyethanol mole fraction. It can be seen from the Fig. 1 that the logarithmic excess viscosities are positive over the entire range of mole fraction both in 2-butoxyethanol–n-hexane and -n-heptane mixtures at the two different temperatures. The rise in temperature was observed to influence differently in n-hexane and -n-heptane mixtures at about equimolar concentrations. A slight decrease was noted in the former and an increase was observed in the later.

The free energy of activation of flow, ΔG^E derived from experimental viscosities and also calculated from Eq. 12 using the constants from the Table 4 are shown in Fig. 2. The ΔG^E was found to be positive in both the mixtures and follow the similar pattern as logarithmic excess viscosities.

As far as we are aware, there is no viscosity data on the similar systems in the literature for a comparison. In order to ascertain the dominant thermodynamic factor that contribute to the present mixture viscosity and for explaining our results, we have evaluated the theoretical viscosities of the mixtures through Flory theory and following an approach proposed by Bloomfield and Dewan. The pertinent relations that are used are already given in Eqs. 2–5.

The various individual contributions viz. enthalpy, entropy, free energy and free volume viscosities together with their combinations to the overall mixture viscosity are given in Table 5. An inspection of the data reveals that the contribution $\eta_{id}\ \eta_S$ is

Table 4 The coefficients of Eq. 12 for the excess viscosity functions of the liquid mixtures.

	303.15 K		*313.15 K*	
	$\Delta ln\eta^E$, *cP*	ΔG^E, *J.mol*$^{-1}$	$\Delta ln\eta^E$, *cP*	ΔG^E, *J.mol*$^{-1}$
	2-Butoxyethanol–n-hexane			
a_0	0.5328	1636.6	0.7383	1812.3
a_1	0.0225	−98.0	−0.3513	−215.4
a_2	0.4554	240.7	−0.1020	−484.3
a_3	0.3577	−379.1	1.6090	541.5
a_4				563.0
σ	0	0	0.0003	0
	2-Butoxyethanol–n-hexane			
a_0	0.8115	2127	0.6423	1917.4
a_1	0.0333	−12.1	0.0652	−47.2
a_2	−0.2807	−605.5	−0.1863	−721.5
a_3	0.1962	−483.6	−0.1754	−205.3
σ	0	0	0.0003	0.008

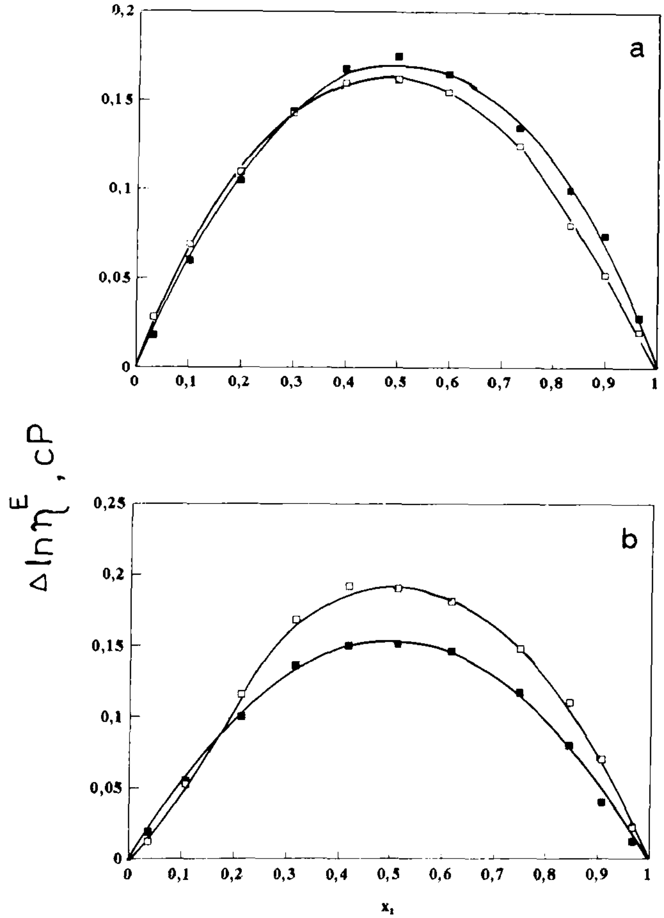

Figure 1 Excess viscosities for a) 2-butoxyethanol n-hexane and b) 2-butoxyethanol n-heptane mixtures at (■) 303.15 and (□) 313.15 K.

found to be close to experimental viscosities in case of both the mixtures. The product $\eta_{id}\ \eta_S$ in both the mixtures was slightly more than η_{id} whereas other combinations, in general are smaller than the ideal viscosity. The enthalpies of 2-butoxyethanol–n-heptane mixtures at 25°C^2 were reported to be positive over the entire range of butoxyethanol concentration. If the same trend is extrapolated to our mixtures at 30°C, the smaller values of $\eta_{id}\eta_H$ over η_{id} can be expected. The small value of the combination $\eta_{id}\ \eta_V$ in both the mixtures indicate that the free volume of the mixutre is smaller than the pure components. However the higher values of $\eta_{id}\eta_S$ over η_{id} emphasize the changes in the orderness of the molecules upon mixing.

The difference in the logarithmic excess viscosities calculated theoretically from Eq. 3 and those derived from experimentally measured viscosities from Eq. 9 presented further insight into the nature of interactions in the mixtures under study. The deviations in excess viscosities for both the mixtures are given in Table 6 at 303.15 K. It can be seen from the Table 6 that the individual enthalpy contributions

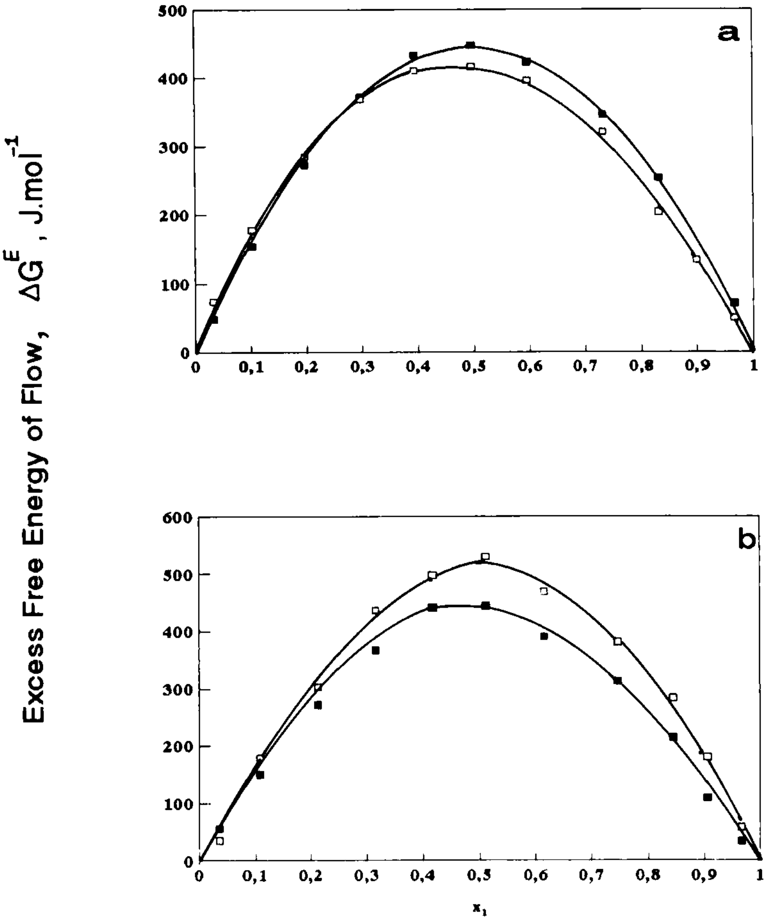

Figure 2 Excess free energy of flow for a) 2-butoxyethanol–n-hexane and b) 2-butoxyethanol–n-heptane mixtures at (■) 303.15 and (□) 313.15 K.

and also the combination of free energy and free volume viscosity contributions at equimolar concentrations are close to the theoretical excess viscosity. Whereas the experimental excess viscosities are not even in sign agreement with $\ln \eta_H$ and $\ln \eta_G \eta_V$ values. The entropy contribution was however positive and seems to be dominant factor for the observed experimental logarithmic excess viscosities. The high positive deviation in the experimental and calculated excess viscosities are indicative of the presence of nonspecific disorder or deorientation interactions as suggested by Delmas et al.[23] between 2-butoxyethanol and n-hexane and n-heptane molecules. The positive excess viscosities as presented in Fig. 1 are also supportive to this conclusion. Similar positive deviations were also attributed to the presence of disorder interactions in case of ethyl acetate–hexane[24] and cyclohexene–organic solvents[25] mixtures.

An attempt is also made to derive the mixture viscosities following the Grunberg-Nissan and Mc Allister approaches and the related equations for the same are

Table 5 Experimental and calculated viscosities of butoxyethanol-alkane mixtures at 303.15 K.

x_1	η_{exptl}	η_{id}	η_H	η_S	η_G	η_V	$\eta_{id}\eta_H$	$\eta_{id}\eta_G$	$\eta_{id}\eta_S$	$\eta_{id}\eta_V$	$\eta_{id}\eta_H\eta_V$	$\eta_{id}\eta_H\eta_S\eta_V$
						Butoxyethanol-n-hexane						
0.0327	0.292	0.284	0.927	1.023	0.948	0.975	0.263	0.269	0.291	0.277	0.256	0.262
0.1014	0.354	0.330	0.829	1.038	0.861	0.954	0.274	0.284	0.343	0.315	0.261	0.271
0.1974	0.456	0.408	0.728	1.056	0.769	0.930	0.297	0.314	0.431	0.379	0.276	0.291
0.2974	0.587	0.509	0.651	1.085	0.706	0.898	0.331	0.359	0.552	0.457	0.297	0.322
0.3950	0.741	0.631	0.623	1.077	0.676	0.899	0.393	0.427	0.680	0.567	0.353	0.380
0.4968	0.930	0.790	0.610	1.079	0.657	0.895	0.482	0.519	0.852	0.707	0.431	0.465
0.5957	1.148	0.984	0.622	1.074	0.668	0.897	0.612	0.657	1.057	0.883	0.549	0.589
0.7323	1.507	1.330	0.679	1.059	0.719	0.914	0.903	0.956	1.408	1.216	0.825	0.874
0.8316	1.793	1.655	0.758	1.037	0.786	0.937	1.254	1.301	1.716	1.551	1.175	1.218
0.8992	2.025	1.922	0.836	1.026	0.858	0.959	1.607	1.649	1.972	1.843	1.541	1.581
0.9658	2.271	2.226	0.940	1.006	0.946	0.988	2.092	2.106	2.239	2.199	2.067	2.079
						Butoxyethanol-n-heptane						
0.0365	0.407	0.403	0.942	1.024	0.965	0.968	0.379	0.388	0.412	0.389	0.366	0.375
0.1077	0.484	0.459	0.879	1.048	0.921	0.946	0.403	0.434	0.481	0.434	0.381	0.399
0.2126	0.626	0.558	1.239	1.069	1.324	0.921	0.691	0.739	0.597	0.514	0.637	0.681
0.3160	0.799	0.676	1.294	1.082	1.400	0.904	0.875	0.946	0.731	0.611	0.791	0.856
0.4170	0.987	0.815	1.363	1.087	1.482	0.895	1.110	1.208	0.886	0.729	0.994	1.080
0.5114	1.177	0.973	1.380	1.089	1.503	0.902	1.343	1.462	1.060	0.878	1.212	1.320
0.6155	1.411	1.177	1.365	1.081	1.476	0.898	1.607	1.737	1.272	1.057	1.443	1.560
0.7469	1.741	1.502	1.291	1.063	1.372	0.916	1.939	2.061	1.597	1.376	1.776	1.888
0.8450	2.011	1.801	1.197	1.041	1.246	0.942	2.156	2.244	1.875	1.697	2.031	2.114
0.9057	2.162	2.016	1.125	1.025	0.153	0.963	2.268	2.324	2.066	1.941	2.184	2.239
0.9663	2.306	2.256	1.049	1.011	1.061	0.984	2.367	2.394	2.281	2.220	2.329	2.355

Table 6 Entrpoy lnη, Enthalpy lnη, Free Energy lnη and Free Volume lnη contributions to the mixture viscosity at equimolar concentrations at 303.15 K.

Butoxy-ethanol+	$\tilde{v}$	$ln\eta_S$	$ln\eta_H$	$ln\eta_G$	$ln\eta_V$	$\Delta ln\eta_{cal}$	$\Delta ln\eta_{exp}$	δ*
Hexane	1.2806	0.0777	−0.5098	−0.4321	−0.1137	−0.5458	0.1527	0.6985
Heptane	1.2785	0.0841	−0.3527	−0.2686	−0.1134	−0.3820	0.0390	0.4210

*$\delta = \Delta\ln\eta_{\mathrm{exp}} - \Delta\ln\eta_{\mathrm{cal}}$

Table 7 Values of the Interaction Parameter (d_{12}), the adjustible parameters v_{ij} and the Average Absolute Deviation (AAD*) for the liquid mixtures at 303.15 and 313.15 K.

				AAD	
T, K	d_{12}*(Eq. 8)*	v_{12}*(Eq. 9)*	v_{21}*(Eq. 9)*	*Eq. 8*	*Eq. 9*
		Butoxyethanol-n-hexane			
303.15	0.847	1.4805	0.7762	0.0139	0.002
313.15	0.702	--	--	0.0108	--
		Butoxyethanol-n-heptane			
303.15	0.788	1.7269	0.6869	0.0596	0.015
313.15	0.630			0.0095	--

*AAD = $\Sigma|\eta_{\mathrm{exp}} - \eta_{\mathrm{cal}}|/n$, where n = no. of measurements

already given in Eq. 8 and 9. The adjustible parameters that appear in Eqs. 8 and 9 are derived from the fits of experimental viscosities. These parameters are in turn are substituted in the same equations to calculate the mixture viscosities over the entire range of concentration. The values of the adjustible parameters together with the absolute average deviations (AAD) between the experimental and calculated viscosities are given in Table 7. The very smaller values of AAD indicate that the mixture viscosities can be reasonably represented by both the approaches.

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