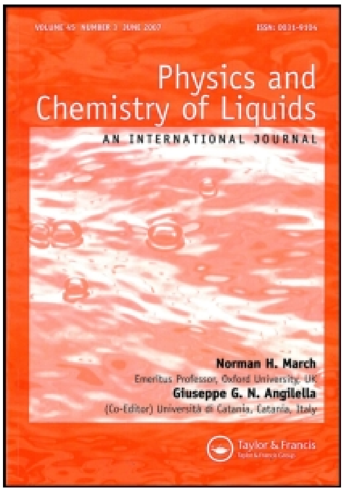

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Viscosimetric and ultrasonic studies of intermolecular interactions of 2-methoxyethanol with diethylene glycol, triethylene glycol and tetraethylene glycol binary mixtures

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Viscosimetric and ultrasonic studies of intermolecular interactions of 2-methoxyethanol with diethylene glycol, triethylene glycol and tetraethylene glycol binary mixtures

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The viscosities at $T = (293.15, 298.15 \text{ and } 303.15)$ K and ultrasonic speeds at $T = 298.15$ K in the binary liquid mixtures of 2-methoxyethanol with diethylene glycol, triethylene glycol and tetraethylene glycol have been measured over the entire mixture compositions. From the experimental data, deviations in the viscosity ($\Delta\eta$), ultrasonic speed (Δu), and excess energies of activation for viscous flow (ΔG^{*E}) have been calculated. The viscosity data were correlated with equations of Hind *et al.*, Grunberg and Nissan, Frenkel, and McAllister. The results are discussed in terms of intermolecular interactions and structure of studied binary mixtures.

Keywords: 2-Methoxyethanol; Ethylene glycols; Viscosity; Speed of sounds

1. Introduction

In our previous papers, we investigated the relative permittivity and volumetric properties of binary mixtures of 2-methoxyethanol (ME) with ethylene glycols [1,2]. Obtained results and literature information about structural properties of ME and glycols [3–7] seem to indicate that the stable intermolecular complexes of the ME·DEG, ME·TEG and ME·TETRAEG types are, respectively, formed in the studied binary mixtures.

Mixtures containing alkoxyethanols and polyethylene glycols are very important from a theoretical point of view, not only because of their self-association, but also due to the strong intermolecular effects produced by presence of –O– and –OH groups in the same molecule [3–7].

In continuation of our program on the thermodynamic, structural and physico-chemical properties of some mixtures of alkoxyalcohols with ethylene glycols, the present article reports the viscosity (η) at $T = (293.15, 298.15 \text{ and } 303.15)$ K, and speed of sound (u) at $T = 298.15$ K for binary mixtures containing 2-methoxyethanol,

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diethylene glycol (DEG), triethylene glycol (TEG) and tetraethylene glycol (TETRAEG). From these results, the deviations of the viscosity ($\Delta\eta$) at $T = (293.15$, 298.15 and 303.15) K, deviations in the speed of sound (Δu), and excess energies of activation for viscous flow (ΔG^{*E}) at $T = 298.15$ K have been calculated. These quantities have been fitted to the Redlich–Kister equation [8], to obtain the binary coefficients and standard deviations. Furthermore, the experimental results have been used to describe the nature of intermolecular interactions.

2. Experimental

2.1. *Materials*

The following materials with mole fraction purity as stated were used: 2-methoxyethanol (Merck – Schuchardt FRG, GC > 0.99 mole fraction), diethylene glycol (Fluka, Switzerland, puriss. p.a., GC $\geq$ 0.995 mole fraction), triethylene glycol (Fluka, Switzerland, puriss. anhydrous, GC > 0.99 mole fraction) and tetraethylene glycol (Fluka, Switzerland, purum, GC $\geq$ 0.99 mole fraction). All glycols and 2-methoxyethanol were further purified by the methods described by Sastry [9], Iglesias [10] and Pal [11].

The mixtures were prepared using a Sartorius balance. Conversion to molar quantities was based on the relative atomic mass table of 1985, issued by IUPAC in 1986. The maximum estimated error in the mole fractions is $\pm 1 \times 10^{-4}$. Liquids were stored in a dry-box over phosphorus pentoxide and degassed by ultrasound just before the experiment.

2.2. *Measurements*

The flow times of the mixtures and the pure liquids were measured in a ViscoClock (made by Schott), equipped with an Ubbelohde capillary viscometer. The viscometer was calibrated with water. The water used was deionized by ion-exchange resins and then doubly distilled. Its conductivity was always lower than $10^{-7}\ \Omega^{-1}\,\mathrm{cm}^{-1}$. The time measurement uncertainty was ±0.005%, and the display resolution ±0.01 s. The uncertainty in the viscosity measurement was estimated to be less than ±0.001 mPa s. All measurements described above were performed at least three times, and the results reported are the averages of these measurements. In all measurements of the viscosity, a Haake model DC-30 thermostat was used that controlled the temperature of the instrument to ±0.01 K.

The speed of sound was measured by resonance method using the ResoScan™ System (Germany) apparatus. The speed of sound are determined from series of resonance frequencies of the resonator calls and also from waviness due to multiple reflections in the signal transmitted through the path length cell [12]. The operating frequency of the transducers was 8.3 MHz. The relative error of the measured speed of sound was lower than 1×10^{-5} over the entire range of concentration. The temperature of the samples was controlled to within ±0.005 K by Peltier thermostat and it was measured to accuracy of ±0.01 K.

3. Results and discussion

The experimental values of viscosities (η) at $T=(293.15$, 298.15 and 303.15) K, and speed of sounds, at $T=298.15$ K, are summarized in tables 1–4.

The deviation of the viscosity from a mole fraction average was calculated from the following equation:

$$\Delta\eta = \eta_2(x_1 \cdot \eta_1 + x_2 \cdot \eta_2), \tag{1}$$

where η_1, η_2 and η are the viscosities of the 2-methoxyethanol, ethylene glycols and the mixtures, respectively. The values of $\Delta\eta$, at temperatures of (293.15, 298.15 and 303.15) K, are summarised in tables 1–3.

The excess energies of activation for viscous flow, at $T=298.15$ K, were obtained with the expression:

$$\Delta G^{*E} = R \cdot T \cdot \left[(\ln\eta \cdot V) - \left[x_1 \cdot (\ln\eta_1 \cdot V_1) + x_2 \cdot (\ln\eta_2 \cdot V_2)\right]\right], \tag{2}$$

where R is gas constant, T is absolute temperature, V_1, V_2 and V are the molar volumes of the 2-methoxyethanol, ethylene glycols and the mixtures, respectively (see [2]).

From the experimental data of speed of sound, the deviations in the speed of sound (Δu) from a mole fraction average were calculated from the following equation:

$$\Delta u = u - (x_1 \cdot u_1 + x_2 \cdot u_2) \tag{3}$$

Table 1. Viscosity (η) and deviations in viscosity ($\Delta\eta$) for 2-methoxyethanol (1) + diethylene glycol (2) binary mixtures at $T=(293.15$, 298.15, and 303.15) K.

ME (1) + DEG (2)						
	η (mPa s)			$\Delta\eta$ (mPa s)		
x_1	293.15 K	298.15 K	303.15 K	293.15 K	298.15 K	303.15 K
0.0000	38.816	29.978	21.875	0.000	0.000	0.000
0.0630	32.909	25.609	18.574	−3.569	−2.578	−2.011
0.1046	29.551	23.079	16.704	−5.384	−3.925	−3.030
0.1548	25.972	20.368	14.706	−7.100	−5.208	−4.000
0.1955	23.411	18.404	13.264	−8.150	−6.015	−4.609
0.2599	19.896	15.69	11.275	−9.276	−6.898	−5.280
0.3032	17.817	14.096	10.112	−9.748	−7.261	−5.556
0.3425	16.134	12.784	9.154	−9.973	−7.455	−5.710
0.3978	14.016	11.129	7.954	−10.038	−7.538	−5.778
0.4463	12.378	9.856	7.030	−9.877	−7.432	−5.709
0.4981	10.818	8.633	6.143	−9.515	−7.182	−5.535
0.5488	9.459	7.565	5.383	−8.992	−6.808	−5.258
0.5957	8.321	6.686	4.755	−8.390	−6.354	−4.925
0.6523	7.085	5.718	4.079	−7.525	−5.713	−4.443
0.6987	6.176	5.003	3.590	−6.713	−5.108	−3.982
0.7511	5.269	4.232	3.094	−5.675	−4.389	−3.405
0.8021	4.393	3.612	2.670	−4.659	−3.559	−2.785
0.8546	3.602	2.998	2.276	−3.502	−2.680	−2.104
0.8964	3.013	2.551	1.995	−2.539	−1.939	−1.530
0.9423	2.414	2.084	1.714	−1.435	−1.101	−0.871
1.0000	1.708	1.544	1.404	0.000	0.000	0.000

Table 2. Viscosity (η) and deviations in viscosity ($\Delta\eta$) for 2-methoxyethanol (1) + triethylene glycol (2) binary mixtures at $T = (293.15, 298.15, \text{and } 303.15)$ K.

ME (1) + TEG (2)						
	η (mPa s)			$\Delta\eta$ (mPa s)		
x_1	293.15 K	298.15 K	303.15 K	293.15 K	298.15 K	303.15 K
0.0000	41.099	34.392	29.244	0.000	0.000	0.000
0.0555	35.941	30.261	26.075	−2.972	−2.308	−1.624
0.1005	32.077	27.247	23.695	−5.063	−3.844	−2.751
0.1526	27.985	24.056	21.134	−7.103	−5.323	−3.862
0.2010	24.535	21.358	18.930	−8.646	−6.432	−4.718
0.2544	21.124	18.653	16.689	−9.954	−7.382	−5.473
0.3016	18.451	16.491	14.872	−10.768	−7.994	−5.975
0.3421	16.375	14.789	13.440	−11.248	−8.366	−6.280
0.4006	13.736	12.583	11.544	−11.583	−8.650	−6.547
0.4493	11.83	10.946	10.132	−11.571	−8.687	−6.603
0.4936	10.297	9.622	8.980	−11.359	−8.556	−6.522
0.5499	8.592	8.124	7.664	−10.846	−8.205	−6.271
0.6007	7.271	6.950	6.621	−10.166	−7.710	−5.900
0.6501	6.157	5.943	5.714	−9.334	−7.095	−5.431
0.7042	5.105	4.973	4.825	−8.255	−6.287	−4.814
0.7531	4.289	4.207	4.115	−7.145	−5.447	−4.163
0.7931	3.714	3.650	3.584	−6.144	−4.690	−3.580
0.8455	3.061	2.991	2.954	−4.733	−3.628	−2.751
0.9009	2.487	2.385	2.346	−3.125	−2.414	−1.817
0.9500	2.069	1.924	1.856	−1.609	−1.262	−0.940
1.0000	1.708	1.544	1.404	0.000	0.000	0.000

Table 3. Viscosity (η) and deviations in viscosity ($\Delta\eta$) for 2-methoxyethanol (1) + tetraethylene glycol (2) binary mixtures at $T = (293.15, 298.15, \text{and } 303.15)$ K.

ME (1) + TETRAEG (2)						
	η (mPa s)			$\Delta\eta$ (mPa s)		
x_1	293.15 K	298.15 K	303.15 K	293.15 K	298.15 K	303.15 K
0.0000	53.236	44.451	35.686	0.000	0.000	0.000
0.0725	46.352	39.141	31.64	−3.148	−2.199	−1.561
0.1073	43.167	36.716	29.69	−4.540	−3.131	−2.318
0.1599	38.535	33.160	26.825	−6.462	−4.430	−3.379
0.2016	35.040	30.448	24.659	−7.808	−5.353	−4.116
0.2489	31.296	27.473	22.298	−9.115	−6.298	−4.855
0.3027	27.332	24.237	19.768	−10.306	−7.226	−5.541
0.3523	23.940	21.441	17.621	−11.143	−7.894	−5.987
0.4136	20.141	18.110	15.099	−11.783	−8.595	−6.408
0.4537	17.874	16.108	13.573	−11.984	−8.876	−6.559
0.4979	15.585	14.049	12.016	−11.995	−9.039	−6.601
0.5484	13.202	11.874	10.275	−11.776	−9.047	−6.611
0.6014	10.996	9.852	8.723	−11.251	−8.795	−6.346
0.6523	9.117	8.152	7.342	−10.507	−8.311	−5.982
0.6991	7.604	6.801	6.195	−9.609	−7.654	−5.524
0.7477	6.243	5.586	5.137	−8.466	−6.783	−4.916
0.7997	4.983	4.506	4.166	−7.046	−5.632	−4.105
0.8523	3.917	3.590	3.323	−5.402	−4.291	−3.144
0.8920	3.214	3.000	2.772	−4.059	−3.178	−2.334
0.9486	2.372	2.235	2.067	−1.985	−1.514	−1.099
1.0000	1.708	1.544	1.404	0.000	0.000	0.000

Table 4. Speed of sound (u) and deviations in speed of sound (Δu) for 2-methoxyethanol (1) + ethylene glycols (2) binary mixtures at $T = 298.15$ K.

ME + DEG			ME + TEG			ME + TETRAEG		
	298.15 K			298.15 K			298.15 K	
x_1	u (m s^{-1})	Δu (m s^{-1})	x_1	u (m s^{-1})	Δu (m s^{-1})	x_1	u (m s^{-1})	Δu (m s^{-1})
0.0000	1579.003	0.000	0.0000	1608.725	0.000	0.0000	1597.865	0.000
0.1006	1558.200	3.017	0.0991	1591.704	9.399	0.0989	1585.491	12.910
0.1996	1537.100	5.371	0.1986	1573.112	17.323	0.2001	1570.157	23.460
0.3006	1514.832	7.031	0.2998	1552.472	23.675	0.3004	1553.652	32.591
0.4130	1489.270	8.079	0.4015	1529.813	28.118	0.3990	1535.428	39.593
0.5001	1469.112	8.548	0.5009	1505.645	30.437	0.4997	1513.862	43.770
0.6007	1445.114	8.373	0.5993	1479.373	30.408	0.6002	1489.357	44.971
0.6989	1421.100	7.636	0.6999	1449.787	27.620	0.6995	1461.330	42.330
0.8004	1395.650	6.226	0.7993	1417.394	21.744	0.7989	1428.607	35.041
0.9003	1369.560	3.773	0.9003	1381.391	12.644	0.9004	1389.484	21.846
1.0000	1342.162	0.000	1.0000	1342.162	0.000	1.0000	1342.162	0.000

The deviations of viscosity, speed of sound and excess energies of activation for viscous were fitted by a Redlich–Kister-type equation [8]:

$$\Delta\eta \,\text{mPa s or } \Delta u \,\text{m s}^{-1}, \text{ and } \Delta G^{*E} \,\text{kJ mol}^{-1} = x_1 \cdot (1 - x_1) \sum_{j=0}^{4} a_j \cdot (2x_1 - 1)^j \quad (4)$$

The parameters a_j of equation (4) were evaluated by the least-squares method. The values of these parameters, at each studied temperature, with standard deviation σ, are summarized in table 5.

The standard deviation values were obtained from:

$$\sigma = \left[\frac{\sum (X_{\text{exptl}} - X_{\text{calcd}})^2}{N - p}\right]^{1/2}, \quad (5)$$

where N is the number of experimental points, $p = 4$ is the number of parameters, X_{exptl} and X_{calcd} are the experimental and calculated properties, respectively.

The variations of $\Delta\eta$, Δu and ΔG^{*E} values as a function of the mole fraction of 2-methoxyethanol (x_1) at one temperature ($T = 298.15$ K) for all studied mixtures are shown in the figures 1–3.

Figure 1 shows that $\Delta\eta$ are negative for all three systems with the minimum lying always near $x_1 \approx 0.40$ for ME + DEG, $x_1 \approx 0.45$ for ME + TEG, and $x_1 \approx 0.50$ for ME + TETRAEG binary mixtures. The value of $\Delta\eta$ becomes more negative as the number of oxyethylene group –O–CH_2–CH_2– in the ethylene glycols increases and temperature decreases (tables 1–3):

$$\left|\Delta\eta_{\min}^{\text{ME+TETRAEG}}\right| > \left|\Delta\eta_{\min}^{\text{ME+TEG}}\right| > \left|\Delta\eta_{\min}^{\text{ME+DEG}}\right|$$

This reveals that the strength of the specific interactions is not the only factor influencing the viscosity deviation of the studied liquid mixtures. The molecular size and shapes of the component are equally important factors. An increment of temperature diminishes the self-association in the pure components and also the

Table 5. Coefficients a_j and standard deviations $\sigma(\Delta\eta)$, $\sigma(\Delta G\eta^{*o})$, and $\sigma(\Delta u)$ of equation (4) for ME + DEG, ME + TEG and ME + TETRAEG binary mixtures.

ME + DEG						
T (K)	a_0	a_1	a_2	a_3	a_4	$\sigma(\Delta\eta)$
293.15	−37.9970	17.7086	−6.3458	2.2010	−1.1767	0.010
298.15	−28.6650	12.5991	−4.5799	1.0314	0.3031	0.009
303.15	−22.0960	9.1262	−3.2802	1.4659	−0.7233	0.003
T (K)	a_0	a_1	a_2	a_3	a_4	$\sigma(\Delta G^{*o})$
298.15	115.1470	92.5706	45.5301	43.7185	41.3809	0.286
T (K)	a_0	a_1	a_2	a_3	a_4	$\sigma(\Delta u)$
298.15	33.9621	3.2852	6.1885	3.2418	−0.3052	0.027
ME + TEG						
T (K)	a_0	a_1	a_2	a_3	a_4	$\sigma(\Delta\eta)$
293.15	−45.2660	14.7105	−1.5074	−2.4939	1.7829	0.003
298.15	−34.1170	9.6628	0.3752	0.0389	−2.2886	0.004
303.15	−26.0110	6.6014	1.7103	−0.5009	−1.1871	0.004
T (K)	a_0	a_1	a_2	a_3	a_4	$\sigma(\Delta G^{*o})$
298.15	140.7240	40.2723	44.1718	7.5991	−34.3492	0.235
T (K)	a_0	a_1	a_2	a_3	a_4	$\sigma(\Delta\eta)$
298.15	121.7537	23.7942	1.5971	−3.0869	0.4644	0.029
ME + TETRAEG						
T (K)	a_0	a_1	a_2	a_3	a_4	$\sigma(\Delta\eta)$
293.15	−47.9860	4.2396	4.3619	−1.3609	1.4494	0.005
298.15	−36.1910	−3.7530	5.7269	5.8835	−0.5169	0.011
303.15	−26.4880	−0.1893	0.7327	0.3465	4.8826	0.018
T (K)	a_0	a_1	a_2	a_3	a_4	$\sigma(\Delta G^{*o})$
298.15	289.7653	29.8091	−31.5055	136.6412	175.9952	0.753
T (K)	a_0	a_1	a_2	a_3	a_4	$\sigma(\Delta\eta)$
298.15	175.6286	56.3144	9.9874	8.8832	28.7024	0.068

hetroassociation between unlike molecules, because of the increase of the thermal energy. This leads to less negative values of $\Delta\eta$ temperature is raised as observed in the present study. Similar conclusions we reported earlier from the analysis, the volumetric and dielectric properties of binary mixtures of 2-methoxyethanol with ethylene glycols [1,2].

As suggested by other authors [13–16], the study of this structural parameter ($\Delta\eta$) for binary liquid systems represents a unique tool for investigating the formation of intermolecular complexes, and provides a valuable aid for determining their stoichiometry. The position of the relative minima or maxima in the plots of $\Delta\eta$ *versus* x_1, could be taken as the true composition of these intermolecular complexes.

Alkoxyethanols and ethylene glycols are a very interesting class of the solvents, due to the simultaneous presence of the oxy and hydroxyl groups in the same molecule, which allow their self-association via intra- and/or intermolecular hydrogen bonds and by dipole–dipole interactions. The formation of stable intermolecular hydrogen bonds is more favorable when the molecules are in the *gauche* conformations [3–7]. The addition of pure ME to ethylene glycols (or ethylene glycols to ME) would disrupt their self-associated internal structure, followed by hydrogen bonding or/and dipolar interactions between unlike molecules.

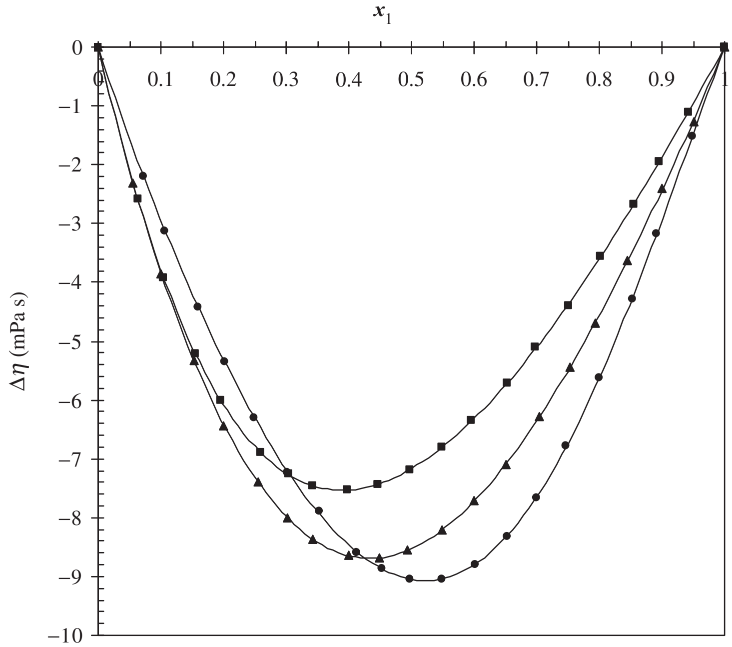

Figure 1. Plot of deviations in the viscosity ($\Delta\eta$) against mole fraction ME (x_1) for {(■) ME (1) + DEG (2), (▲) ME (1) + TEG (2) and (●) ME (1) + TETRAEG (2)} binary liquid mixtures, at $T = 298.15$ K.

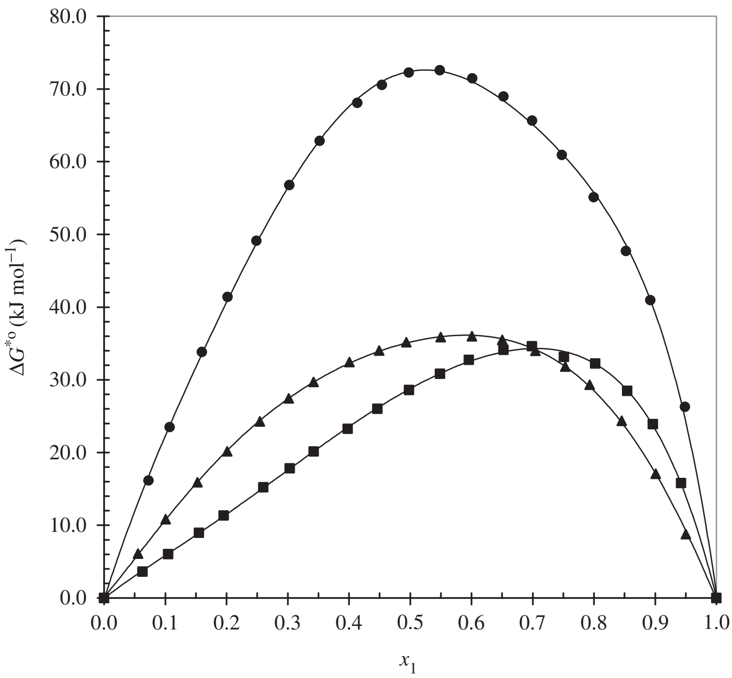

Figure 2. Plot of excess energies of activation for viscous flow (ΔG^{*o}) against mole fraction ME (x_1) for {(■) ME (1) + DEG (2), (▲) ME (1) + TEG (2) and (●) ME (1) + TETRAEG (2)} binary liquid mixtures, at $T = 298.15$ K.

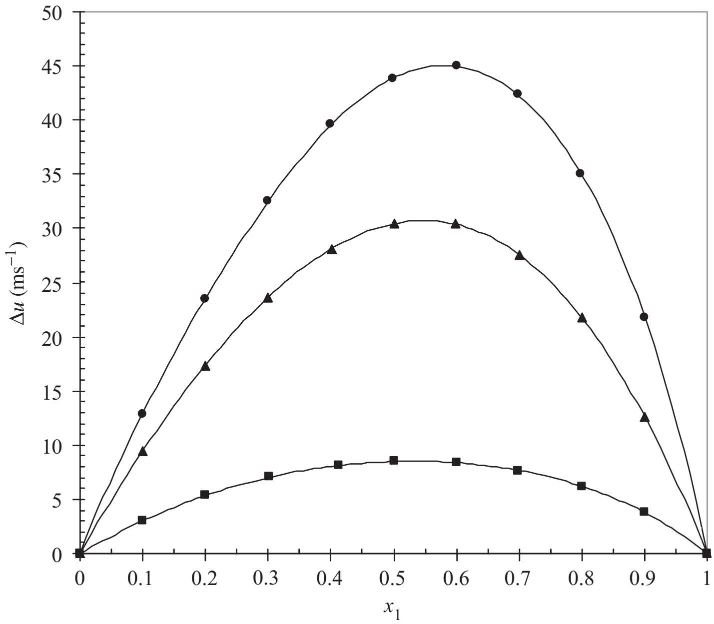

Figure 3. Plot of deviations in the speed of sound (Δu) against mole fraction ME (x_1) for {(■) ME (1) + DEG (2), (▲) ME (1) + TEG (2) and (•) ME (1) + TETRAEG (2)} binary liquid mixtures, at $T = 298.15$ K.

Similar conclusions can be drawn from the analysis of the excess energies of activation for viscous flow (ΔG^{*E}) and deviation of the speeds of sound (Δu) (figures 2 and 3).

Figures 2 and 3 shows that ΔG^{*E} and Δu are positive for all three systems. The maximum of ΔG^{*E} lying always near $x_1 \approx 0.55$ for ME + DEG, $x_1 \approx 0.60$ ME + TEG and $x_1 \approx 0.70$ ME + TETRAEG binary mixtures, and the maximum of Δu lying always near $x_1 \approx 0.50$ for ME + DEG, $x_1 \approx 0.55$ ME + TEG and $x_1 \approx 0.60$ ME + TETRAEG binary mixtures. The values of ΔG^{*E} and Δu becomes more positive as the number of oxyethylene group –O–CH_2–CH_2– in the ethylene glycols increases:

$$\Delta G^{*E}_{\max} \text{ or } \Delta u_{\max}[\text{ME} + \text{TETRAEG}] > \Delta G^{*E}_{\max} \text{ or } \Delta u_{\max}[\text{ME} + \text{TEG}] > \Delta G^{*E}_{\max} \text{ or } \Delta u_{\max}[\text{ME} + \text{TETRAEG}]$$

As it is suggested by other authors, positive deviations of ΔG^{*E} and Δu should be observed in binary mixtures where hydrogen bonding interactions between unlike molecules take place. In this case, these hydrogen bonding interactions should be predominating over all other interactions (e.g. dispersion forces), which generally are responsible for negative excess in the analysed property [16–24].

Several semi-empirical equations have been used to estimate the viscosity of liquid mixtures in terms of pure component data. The experimental viscosity data of analyzed binary liquid mixtures were further fitted to:

- the Hind *et al.* equation [25]:

$$\eta = x_1^2 \cdot \eta_1 + x_2^2 + 2 \cdot x_1 \cdot x_2 \cdot H_{12}, \tag{6}$$

Table 6. Adjustable parameters and standard deviations of several semiempirical equations for (ME + DEG), (ME + TEG), and (ME + TETRAEG) binary mixtures, at $T = 298.15$ K.

Equation	T (K)	Values of the parameters		σ (%)
ME + DEG				
Hind *et al.*	298.15	$H_{12} = 0.7834$		0.99
Grunberg–Nissan	298.15	$G_{12} = 1.1003$		0.21
Frenkel	298.15	$F_{12} = 12.2775$		1.07
McAllister	298.15	$Z_{12} = 7.6493$	$Z_{21} = 10.7903$	0.99
ME + TEG				
Hind *et al.*	298.15	$H_{12} = 0.7810$		0.73
Grunberg–Nissan	298.15	$G_{12} = 1.0914$		0.31
Frenkel	298.15	$F_{12} = 12.6298$		0.37
McAllister	298.15	$Z_{12} = 9.0848$	$Z_{21} = 11.8403$	0.74
ME + TETRAEG				
Hind *et al.*	298.15	$H_{12} = 5.7066$		0.29
Grunberg–Nissan	298.15	$G_{12} = 2.2421$		0.14
Frenkel	298.15	$F_{12} = 26.5723$		1.44
McAllister	298.15	$Z_{12} = 15.3544$	$Z_{21} = 18.3697$	1.02

- the Grunberg and Nissan equation [26]:

$$\eta = \exp(x_1 \cdot \ln \eta_1 + x_2 \cdot \ln \eta_2 + x_1 \cdot x_2 \cdot G_{12}), \tag{7}$$

where G_{12} is a parameter proportional to the interaction energy.

- the Frenkel equation [27]:

$$\ln \eta = \sum_{i=1}^{n} x_i^2 \cdot \eta_i + 2 \cdot \left[\sum_{i=1}^{n} \sum_{j>1}^{n} x_i \cdot x_j \ln F_{ij} \right], \tag{8}$$

where F_{21} are the parameters representing binary 12 interactions;

- the two parameters McAllister equation [28]:

$$\ln \upsilon = x_1^3 \cdot \ln \eta_1 + 3 \cdot x_1^2 \cdot x_2 \cdot \ln Z_{12} + 3 \cdot x_1 \cdot x_2^2 \cdot \ln Z_{21} + x_2^3 \cdot \ln \upsilon_2 - \ln\left(x_1 + x_2 \cdot \frac{M_2}{M_1} \right) + 3 \cdot x_1^2 \cdot x_2 \cdot \ln\left(\frac{2}{3} + \frac{M_2}{3M_1} \right) + 3 \cdot x_1 \cdot x_2^2 \cdot \ln\left(\frac{1}{3} + \frac{2M_2}{3M_1} \right) + x_2^3 \cdot \ln\left(\frac{M_2}{M_1} \right) \tag{9}$$

where Z_{12} and Z_{21} are interactions parameters; M_1 and M_2 are the molecular mass; υ_1 and υ_2 are the kinematics viscosity of pure component 1 and 2.

To perform a numerical comparison of the correlating ability of equations (6)–(9), we calculated the standard deviations (σ). The values of the parameters of equations (6)–(9) were determined for the systems using a least-squares method, with equal weights assigned to each experimental datum. The correlation parameters and standard deviations (σ) for these equations are listed in table 6. It is observed that the Grunberg–Nissan relation fit the experimental results better as compared to the Hind, Frenkel and McAllister equations, as the σ values for the latter equations are larger than the values for the Grunberg–Nissan equation in the studied binary mixtures.

Obtained, in this paper, results and literature informations about structural properties of ME and glycols [3–7,29] seem to indicate that the stable intermolecular complexes, respectively, formed by hydrogen bonding and dipolar interactions between unlike molecules in the studied binary mixtures.

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