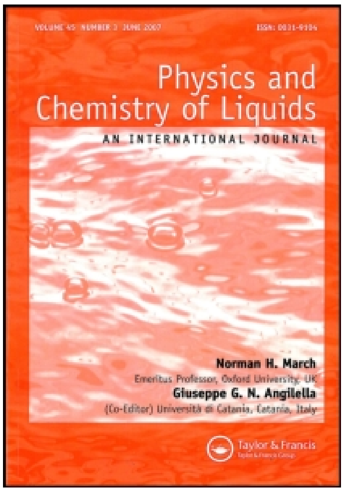

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Volumetric, viscosimetric and acoustic studies of binary mixtures of 2-ethoxyethanol with 1-alkanols at 298.15 K

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Volumetric, viscosimetric and acoustic studies of binary mixtures of 2-ethoxyethanol with 1-alkanols at 298.15 K

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Densities, viscosities and sound speeds of eight binary mixtures of 2-ethoxyethanol with methanol, ethanol, 1-propanol, 1-butanol, 1-pentanol, 1-hexanol, 1-heptanol and 1-octanol are determined over the entire range of mole fractions at 298.15 K. The excess molar volume (V^E), viscosity deviations ($\Delta\eta$) and Gibbs excess free energy of activation for viscous flow (ΔG^{*E}) have been investigated from these experimental density (ρ) and viscosity (η) values. The viscosity data have been correlated by Grunberg and Nissan, Tamura–Kurata and Hind correlation equation. The deviations in isentropic compressibility (ΔK_S) were also calculated using the measured speeds of sound. The results are discussed and interpreted in terms of molecular package and specific interaction predominated by hydrogen bonding. The sound speeds were predicted by using free length and collision factor theoretical formulations, by Nomotto equation, by Vandeal Vangael ideal mixing relation and the impedance dependence relation. The deviations have been fitted to a Redlich–Kister equation and the results are discussed in terms of molecular interactions and structural effects. The excess properties are found to be either negative or positive depending on the molecular interactions and the nature of liquid mixtures.

Keywords: Densities; Viscosities; Excess molar volume; Viscosity deviation; Excess Gibbs energy of activation of viscous flow; 2-Ethoxyethanol; Monoalcohols; Sound speed; Isentropic compressibility; Excess isentropic compressibility; Hydrogen bonding; Specific interactions

1. Introduction

The volumetric and viscometric properties of mixed solvent systems and their dependence on composition find applications in many important chemical, industrial and biological processes. Such systems find industrial applications such as heat transfer, fluid flow and so forth. The study of quantities such as excess molar volume, deviation in viscosity, excess free energy of activation of viscous flow, excess isentropic compressibility, etc. of binary liquid mixtures are useful in understanding the nature and strength of molecular interactions between the component molecules [1–3].

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The present work deals with the study of excess thermodynamic and transport properties of some non-aqueous binary liquid mixtures. The liquids under investigation have been chosen on the basis of their industrial applications. The alkoxyethanols and alkanols are good industrial solvents which occupy an important place in many industrial processes such as pharmaceutical and cosmetics industry. These applications have greatly stimulated the need for extensive information on the thermodynamic, acoustic and transport properties of these solvents and their mixtures [4–9].

In this article, we extend our studies to the binary mixtures formed by 2-ethoxyethanol represented as (**1**) with eight monoalcohols, represented as (**2**), including methanol, ethanol, 1-propanol, 1-butanol, 1-pentanol, 1-hexanol, 1-heptanol and 1-octanol at 298.15 K. The various thermodynamic properties such as excess molar volume (V^E), viscosity deviations ($\Delta\eta$) and Gibbs excess free energy of activation for viscous flow (ΔG^{*E}) obtained from experimental observations have been rationalized. 2-Ethoxyethanol and the monoalcohols have both a proton donor and a proton acceptor group. It is expected that there will be a significant degree of H-bonding leading to self-association in pure state in addition to mutual association in their binaries [10].

Besides this, isentropic compressibilities as well as deviations in isentropic compressibility (ΔK_S) calculated from measured speed of sound (u) of 2-ethoxyethanol and monoalcohol mixtures are presented at 298.15 K. The measurement of ultrasonic speed enables the accurate determination of isentropic compressibility coefficients (K_S) which can be used to provide qualitative information about the physical nature of the aggregates occurring in the liquid phase. To investigate the nature of the interactions, various thermodynamic parameters such as specific acoustic impedance [11] Z, intermolecular free length L_f [12], van der Waal's constant b [13], molecular radius r [12], geometrical volume B, molar surface area Y, available volume V_a [12], molar speed of sound R [14], collision factor S [15] and molecular association M_A [16] has been calculated using the sound speed and density of the mixtures and pure solvents which are sensitive to interaction between solute and solvent. These properties for the mixtures studied are presented, compared and analyzed. A comparative study of the sound speeds calculated using different theoretical formulations and equations with the experimentally measured value is also performed, the results are represented graphically and the standard deviation in each case has been reported.

2. Experiments

2.1. *Source and purity of samples*

2-Ethoxyethanol (Merck, India) was kept several days over anhydrous $CaSO_4$, refluxed for 4 h over CaO. Finally, it was distilled at low pressure. Details have been described elsewhere [17]. Methanol, ethanol, propanol, 1-butanol, 1-pentanol, 1-hexanol, 1-heptanol and 1-octanol (S.D. Fine Chemicals, Analytical Reagent, Purity >99%) were used. The liquids were stored over molecular sieves. The density, viscosity and the sound speeds of the mixtures were determined immediately after mixing. The purity of the solvents was ascertained by GLC and also by comparing experimental values of densities and viscosities with those reported in the literature as listed in table 1.

Table 1. Comparison of density (ρ), viscosity (η) and sound speeds (u) with literature data at 298.15 K.

	$\rho \times 10^{-3}$ (kg m^{-3})		η (mPa s)		u (m s^{-1})	
Pure liquid	Expt.	Lit.	Expt.	Lit.	Expt.	Lit.
2-Ethoxyethanol	0.9254	0.9253 [44]	1.85061	1.851 [60]	1309.00	1300.40 [51]
Methanol	0.7866	0.78656 [45]	0.5509	0.5422 [45]	1104.2	1103.00 [52]
Ethanol	0.7857	0.7851 [46]	1.0892	1.088 [46]	1142	1145.00 [53]
1-Propanol	0.7993	0.7995 [47]	2.0074	2.004 [49]	1209.4	1206.50 [47]
1-Butanol	0.8062	0.8058 [48]	2.5571	2.5600 [49]	1240.2	1240.00 [54]
1-Pentanol	0.8112	0.8111 [48]	3.5104	3.510 [50]	1277.2	1277.00 [54]
1-Hexanol	0.8151	0.81515 [48]	4.5917	4.590 [50]	1328.3	1328.00 [56]
1-Heptanol	0.82068	0.81870 [59]	5.93678	–	1331.5	1330.00 [54]
1-Octanol	0.8218	0.8216 [48]	7.3646	7.363 [50]	1347.7	1347.40 [55]

2.2. *Method*

Viscosities (η) have been measured at 298.15 K, by means of a suspended Ubbelohde-type viscometer [18] which was calibrated at the experimental temperature. The flow times were accurate to ±0.1 s, and the uncertainty in the viscosity measurements, based on our work on several pure liquids, was $\pm 2 \times 10^{-4}$ mPa s. Densities (ρ) were measured at the mentioned temperature with an Ostwald–Sprengel-type pycnometer having bulb volume of about 25 cm^3 and an internal diameter of the capillary of about 1 mm. The pycnometer with the test solution was equilibrated in a thermostatic water bath maintained at ±0.01 K of the desired temperature. The evaporation losses remained insignificant during the time of actual measurements. Averages of triplicate measurements were taken into account. The density values were reproducible to $\pm 3 \times 10^{-5}$ g cm^{-3}. The measurements were done in a thermostatic bath controlled to ±0.01 K. The details of the methods and techniques for determination of these parameters have been described in earlier papers [4–6,19,20]. Speeds of sound were determined by a multifrequency ultrasonic interferometer (Mittal Enterprise, New Delhi) working at 5 MHz. The meter was calibrated with water, methanol, and benzene at 298.15 K. The precisions of the speed of sound measurements are ±0.2 m s^{-1}. The details of the methods and techniques have been described previously [19,20].

The mixtures were prepared by mixing known volumes of pure liquids in airtight stoppered bottles. The reproducibility in mole fraction was within ±0.0002 units. The weights were taken on a Mettler electronic analytical balance (AG 285, made in Switzerland) accurate to 0.02 mg.

3. Results and discussion

The experimentally measured densities (ρ) and viscosities (η) of the pure liquids at 298.15 K along with the reference values are recorded in table 1.

The values of experimental density (ρ) and viscosity (η), the excess properties, (V^E, $\Delta\eta$ and ΔG^{*E}) along with the interaction parameters (d_{12}, T_{12} and H_{12}) of the studied mixtures are recorded in table 2 and the corresponding graphs for V^E and $\Delta\eta$ against the mole fractions of 2-ethoxyethanol (x_1) are marked as figures 1 and 2, respectively.

Table 2. Values of density (ρ), viscosity (η), excess molar volume (V^E), viscosity deviation ($\Delta\eta$), excess Gibbs energy of activation for viscous flow (ΔG^{*E}), Grunberg–Nissan, Tamura Kurata and Hind interaction parameters (d_{12}, T_{12}, H_{12}) for binary mixtures of 2-Ethoxy ethanol+Methanol, Ethanol, Propanol, Butanol, Pentanol, Hexanol, Heptanol and Octanol at 298.15 K.

x_1	$\rho \times 10^{-3}$ (kg m^{-3})	η (mPa s)	$V^E \times 10^6$ (m^3 mol^{-1})	$\Delta\eta$ (mPa s)	ΔG^{*E} (J mol^{-1})	d_{12}	T_{12}	H_{12}
2-Ethoxyethanol + methanol								
0	0.7866	0.5509	0	0	0			
0.0380	0.7997	0.6216	−0.058	0.021	227.32	1.66	1.22	1.49
0.0816	0.8131	0.7062	−0.118	0.049	454.06	1.81	1.26	1.53
0.1322	0.8267	0.7916	−0.176	0.069	625.16	1.65	1.25	1.50
0.1916	0.8406	0.8844	−0.231	0.085	758.35	1.48	1.25	1.47
0.2623	0.8550	0.9885	−0.298	0.097	852.25	1.32	1.24	1.45
0.3478	0.8694	1.1144	−0.349	0.111	914.00	1.21	1.26	1.45
0.4534	0.8839	1.2555	−0.379	0.115	898.30	1.07	1.27	1.43
0.5871	0.8981	1.4225	−0.350	0.109	787.40	0.95	1.29	1.42
0.7619	0.9117	1.6248	−0.211	0.084	531.16	0.85	1.33	1.43
1	0.9254	1.8506	0	0	0			
2-Ethoxyethanol + ethanol								
0	0.7857	1.0892	0	0	0			
0.0538	0.7985	1.1465	−0.058	0.016	72.83	0.45	1.55	1.63
0.1133	0.8115	1.2067	−0.104	0.031	137.4	0.42	1.55	1.63
0.1798	0.8247	1.2721	−0.135	0.046	195.57	0.41	1.56	1.63
0.2542	0.8382	1.3413	−0.165	0.059	241.19	0.39	1.56	1.62
0.3383	0.8521	1.4165	−0.186	0.070	274.98	0.37	1.57	1.63
0.4341	0.8662	1.4966	−0.194	0.077	290.16	0.36	1.58	1.63
0.5440	0.8806	1.5810	−0.185	0.078	280.19	0.34	1.58	1.63
0.6716	0.8953	1.6690	−0.159	0.068	236.54	0.32	1.59	1.63
0.8215	0.9103	1.7613	−0.099	0.047	150.98	0.31	1.61	1.63
1	0.9254	1.8506	0	0	0			
2-Ethoxyethanol + propanol								
0	0.7993	2.0074	0	0	0			
0.0690	0.8108	1.9415	−0.037	−0.047	−64.37	−0.43	1.57	1.50
0.1429	0.8224	1.8906	−0.066	−0.094	−111.04	−0.39	1.60	1.54
0.2223	0.8344	1.8521	−0.100	−0.121	−142.92	−0.36	1.63	1.58
0.3078	0.8466	1.8256	−0.114	−0.134	−158.58	−0.33	1.65	1.62
0.4001	0.8591	1.8108	−0.126	−0.135	−158.41	−0.30	1.68	1.65
0.5001	0.8717	1.8056	−0.113	−0.123	−144.40	−0.26	1.71	1.68
0.6088	0.8844	1.8105	−0.075	−0.102	−116.02	−0.23	1.73	1.72
0.7273	0.8976	1.8226	−0.049	−0.071	−78.28	−0.19	1.76	1.75
0.8572	0.9113	1.8386	−0.022	−0.034	−35.95	−0.15	1.80	1.79
1	0.9254	1.8506	0	0	0			
2-Ethoxyethanol + butanol								
0	0.8062	2.5571	0	0	0			
0.0837	0.8166	2.4339	0.011	−0.064	−54.64	−0.29	1.81	1.79
0.1706	0.8272	2.3117	0.030	−0.125	−112.00	−0.32	1.78	1.76
0.2606	0.8381	2.2060	0.054	−0.167	−154.92	−0.33	1.79	1.77
0.3541	0.8493	2.1129	0.076	−0.194	−186.25	−0.33	1.79	1.78
0.4513	0.8609	2.0349	0.088	−0.203	−201.16	−0.33	1.81	1.79
0.5523	0.8729	1.9838	0.087	−0.183	−183.33	−0.30	1.84	1.83
0.6574	0.8854	1.9489	0.074	−0.144	−143.54	−0.26	1.89	1.88
0.7669	0.8984	1.9249	0.048	−0.090	−87.34	−0.20	1.96	1.95
0.8810	0.9117	1.9016	0.019	−0.033	−27.22	−0.11	2.05	2.05
1	0.9254	1.8506	0	0	0			

(*Continued*)

Table 2. Continued.

x_1	$\rho \times 10^{-3}$ (kg m^{-3})	η (mPa s)	$V^{\mathrm{E}} \times 10^6$ (m^3 mol^{-1})	$\Delta\eta$ (mPa s)	$\Delta G^{*\mathrm{E}}$ (J mol^{-1})	d_{12}	T_{12}	H_{12}
2-Ethoxyethanol + pentanol								
0	0.8112	3.5104	0	0	0			
0.0980	0.8210	3.1674	0.045	−0.180	−54.64	−0.45	1.57	1.66
0.1965	0.8309	2.8637	0.103	−0.321	−112.00	−0.50	1.59	1.67
0.2954	0.8412	2.6191	0.148	−0.401	−154.92	−0.50	1.65	1.72
0.3947	0.8518	2.4305	0.187	−0.425	−186.25	−0.48	1.74	1.79
0.4945	0.8628	2.2827	0.212	−0.407	−201.16	−0.46	1.82	1.87
0.5947	0.8742	2.1783	0.222	−0.345	−183.33	−0.40	1.93	1.96
0.6953	0.8861	2.0992	0.203	−0.257	−143.54	−0.33	2.04	2.07
0.7964	0.8986	2.0275	0.161	−0.161	−87.34	−0.24	2.16	2.18
0.8980	0.9117	1.9541	0.093	−0.066	−27.22	−0.12	2.29	2.32
1	0.9254	1.8506	0	0	0			
2-Ethoxyethanol + hexanol								
0	0.8151	4.5917	0	0	0			
0.1119	0.8244	3.9460	0.080	−0.339	−114.65	−0.50	2.10	1.52
0.2209	0.8340	3.4770	0.147	−0.509	−175.78	−0.45	2.08	1.74
0.3270	0.8440	3.0803	0.191	−0.615	−232.06	−0.46	2.04	1.82
0.4305	0.8543	2.7757	0.221	−0.636	−254.08	−0.46	2.01	1.92
0.5314	0.8649	2.5612	0.240	−0.574	−225.06	−0.40	1.97	2.07
0.6297	0.8758	2.3806	0.254	−0.485	−185.27	−0.36	1.93	2.18
0.7257	0.8873	2.2425	0.237	−0.360	−119.92	−0.29	1.89	2.32
0.8193	0.8992	2.1213	0.22	−0.225	−51.23	−0.19	1.85	2.46
0.9108	0.9119	2.0053	0.126	−0.090	7.83	−0.40	1.81	2.67
1	0.9254	1.8506	0	0	0			
2-Ethoxyethanol + heptanol								
0	0.8207	5.9368	0	0	0			
0.1253	0.8277	4.9665	0.125	−0.435	−55.68	−0.30	1.16	1.80
0.2438	0.8367	4.2073	0.238	−0.712	−110.60	−0.33	1.36	1.90
0.3559	0.8464	3.6548	0.320	−0.811	−126.78	−0.31	1.64	2.08
0.4623	0.8561	3.2302	0.377	−0.818	−120.28	−0.28	1.88	2.25
0.5632	0.8661	2.9021	0.414	−0.733	−92.86	−0.24	2.10	2.40
0.6592	0.8771	2.6169	0.410	−0.435	−75.95	−0.23	1.16	2.50
0.7505	0.8882	2.3828	0.372	−0.487	−51.16	−0.20	2.38	2.59
0.8376	0.8995	2.1996	0.296	−0.315	−7.38	−0.12	2.55	2.74
0.9207	0.9118	2.0410	0.168	−0.134	32.75	0.07	2.79	2.98
1	0.9254	1.8506	0	0	0			
2-Ethoxyethanol + octanol								
0	0.8218	7.3646	0	0	0			
0.1384	0.8302	6.0618	0.169	−0.540	24.99	−0.03	1.33	2.34
0.2654	0.8389	5.0613	0.289	−0.840	36.69	−0.04	1.60	2.45
0.3825	0.8481	4.2677	0.366	−0.988	29.89	−0.07	1.79	2.52
0.4907	0.8576	3.6490	0.409	−1.010	19.57	−0.10	1.98	2.59
0.5910	0.8675	3.1718	0.428	−0.934	16.36	−0.11	2.16	2.68
0.6843	0.8778	2.8054	0.416	−0.786	26.02	−0.09	2.36	2.79
0.7713	0.8887	2.5039	0.370	−0.608	30.75	−0.08	2.52	2.88
0.8525	0.9002	2.2662	0.291	−0.398	45.28	−0.01	2.71	3.03
0.9286	0.9123	2.0779	0.177	−0.166	69.42	0.26	3.04	3.35
1	0.9254	1.8506	0	0	0			

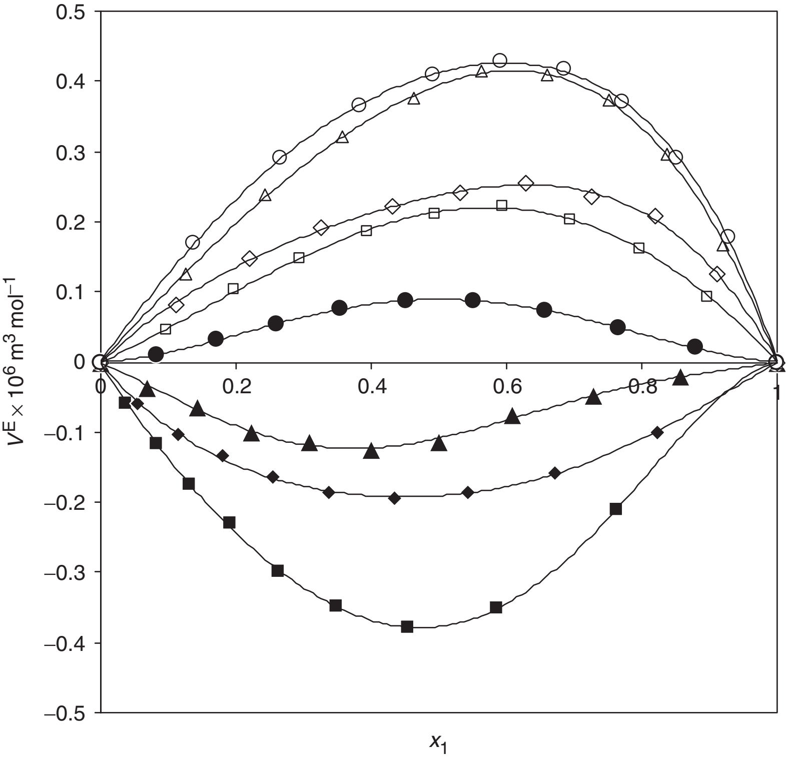

Figure 1. Excess molar volumes (V^E) for binary mixtures of 2-ethoxyethanol with methanol (■), ethanol (◆), 1-propanol (▲), 1-butanol (●), 1-pentanol (□), 1-hexanol (◇), 1-heptanol (△) and 1-octanol (○) at 298.15 K.

3.1. *Excess molar volume*

The excess molar volumes (V^E) were calculated using the molar masses (M_i) and densities of the pure liquids and the mixtures using the following equation [6,23].

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

where, M_i, ρ_i, x_i are the molar mass, density, mole fraction of the ith component and ρ is the density of the mixture, respectively.

From figure 1, we observe that, for the binary mixtures studied here, the V^E values gradually change from highly negative to less negative and finally turn positive with the increase of chain length along the alcohol homologous series. It has highest negative value for methanol and ultimately turns positive for the higher alkanols. V^E values are negative for methanol, ethanol, 1-propanol and then they turn gradually more and more positive from 1-butanol to 1-octanol. Positive V^E values for higher alkanols and negative V^E values for lower alkanols were also reported

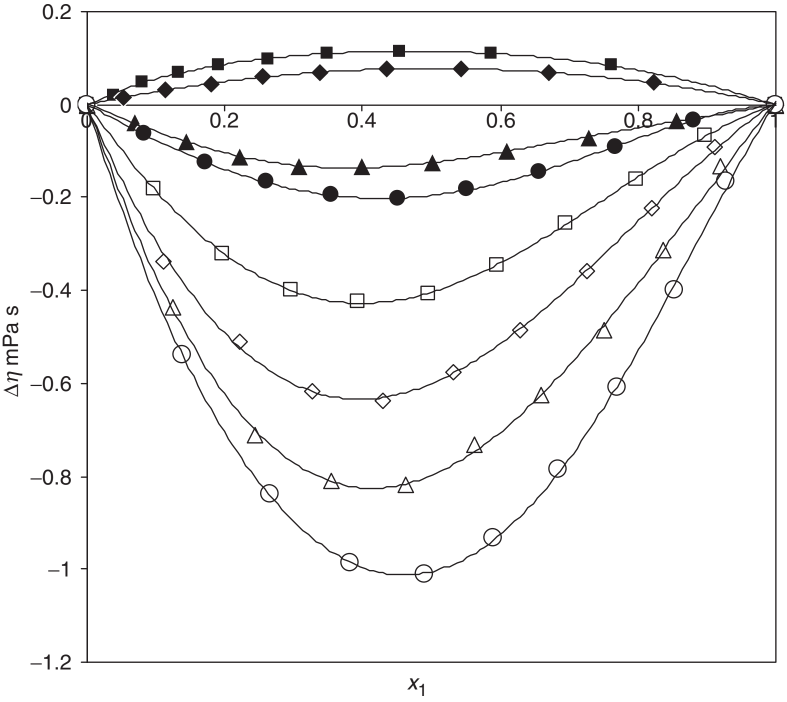

Figure 2. Viscosity deviations (Δη) for binary mixtures of 2-ethoxyethanol with methanol (■), ethanol (◆), 1-propanol (▲), 1-butanol (●), 1-pentanol (□), 1-hexanol (◇), 1-heptanol (Δ) and 1-octanol (○) at 298.15 K.

by some other workers [21–23]. The observed trend in terms of negative values of V^E in 1-alkanol + 2-ethoxyethanol mixture is:

$$\text{Methanol} > \text{Ethanol} > \text{1-Propanol} > \text{1-Butanol} > \text{1-Pentanol}$$
$$> \text{1-Hexanol} > \text{1-Heptanol} > \text{1-Octanol}$$

Such a behavior is the result of the contribution from several contraction and expansion processes which proceed simultaneously when 2-ethoxyethanol-alkan-1-ol molecules are formed. The following effects can be considered: (a) disruption of liquid order on mixing and unfavorable interactions between unlike molecules producing a positive contribution of V^E; (b) contraction due to free volume difference of unlike molecules and (c) possible association through hydrogen bond formation between alkan-1-ol and 2-ethoxyethanol producing a negative contribution to V^E.

The alkanols are known to be extremely self associated through H-bonding and in 2-ethoxyethanol also self association through H-bonding is present [24]. Mixing of 1-alkanols to 2-ethoxyethanol can be expected to bring changes in the H-bonding equilibria and electrostatic interactions with different resultant contributions in the volume of mixing. The negative values obtained for lower 1-alkanols suggest that the interaction between the unlike molecules exceed the structure breaking effect between the like molecules. These interactions are relatively strong between

2-ethoxyethanol and methanol molecules thereby showing the highest negative V^E value for their binary mixture. Increasing the chain length of the alkanols tends to dilute this unlike interaction and finally for the higher alkanols this unlike interaction becomes unfavorable producing a positive contribution of V^E.

3.2. *Viscosity deviations*

The deviation in viscosities from linearity ($\Delta\eta$) can be computed using the relationship,

$$\Delta\eta = \eta - \sum_{i=1}^{j}(x_i\eta_i) \tag{2}$$

The values of $\Delta\eta$ (figure 2) are positive for methanol and ethanol and decrease regularly as the size of the alkan-1-ol is increased. The positive $\Delta\eta$ values indicate the predominance of H-bonding interactions between the unlike molecules over the dissociation effects of the mixing components [25,26]. This results in a liquid structure where the flow is rather difficult than would be expected on the basis of the viscosities of the pure components.

It is known that the strength of the molecular hydrogen bonding is not the only factor influencing the viscosity deviations in liquid mixtures [27,28]. The molecular size and shape of the components and average degree of association of the mixture are equally important factors. The negative values of $\Delta\eta$ for higher alkanols indicate that the average degree of cross-association of mixtures gradually decreases as the chain length of alkan-1-ol is increased [27,29]. Thus, larger the chain length of 1-alkanol, the greater is the decrease in the average degree of association, as a result more negative deviations in viscosity *versus* mole fraction curve are observed. These conclusions are supported by the conclusions drawn from V^E values.

3.3. *Excess Gibbs energies for activation of viscous flow (ΔG^{*E})*

It has been reported that ΔG^{*E} parameter can be considered as a reliable criterion to detect or exclude the presence of interactions between unlike molecules [30,38].

On the basis of the theories of absolute reaction rates [40], the excess Gibbs energy of activation for viscous flow (ΔG^{*E}) was calculated from the equation [41],

$$\Delta G^{*E} = RT\left[\ln\eta V - \sum_{i=1}^{j}(x_i \ln\eta_i V_i)\right] \tag{3}$$

According to Reed and Taylor and Meyer *et al.*, positive ΔG^{*E} values indicate specific interactions while negative values indicate the dominance of dispersion forces [38,39].

The magnitude of the positive value of ΔG^{*E} is an excellent indicator of the strength of specific interactions. From the ΔG^{*E} values recorded in table 2, it is seen that these values are negative or positive keeping similarity with the $\Delta\eta$ values and thereby supports our conclusion drawn from V^E and $\Delta\eta$ considerations.

3.4. *Correlating equations*

Apart from expressing η as a polynomial fit, several semi-empirical relations have been proposed to estimate the dynamic viscosity η of liquid mixtures in terms of pure component data [31,39]. We have examined equations proposed by Grunberg–Nissan, Tamura–Kurata and Hind *et al.*

The single parameter Grunberg–Nissan equation [31] reads as:

$$\eta = \exp\left[\sum_{i=1}^{j}(x_i \ln \eta_i) + d_{12}\prod_{i=1}^{j} x_i\right] \tag{4}$$

where d_{12} is a parameter proportional to the interchange energy and has been regarded as an approximate measure for the non-ideal behaviors of binary mixtures.

Tamura–Kurata [32] put forward the following equation for the viscosity of the binary liquid mixtures:

$$\eta = \sum_{i=1}^{j} x_i\phi_i\eta_i + 2T_{12}\prod_{i=1}^{j}[x_i\phi_i]^{1/2} \tag{5}$$

where T_{12} is the interaction parameter and ϕ_i is the volume fraction of ith pure component in the mixture.

Molecular interactions may also be interpreted by the following viscosity model of Hind *et al.* [33]

$$\eta = \sum x_i^2\eta_i + 2H_{12}\prod_{i=1}^{j} x_i \tag{6}$$

where H_{12} is the interaction parameter.

The interaction parameters have their merits in ascertaining the strength of molecular interactions in binary mixtures. Among the three parameters determined here, the Grunberg–Nissan parameter provides the best measure to ascertain the strength of interaction. At any given composition, the variation of d_{12} with strength of interaction is similar to that of $\Delta\eta$, being negative for systems in which dispersion forces are dominant, becoming less negative and then increasingly positive as the strength of interaction increases [25,34,35,36].

According to Fort and Moore [25], the values of T_{12} and H_{12} are not very different except where the values of the components differ considerably. Furthermore, T_{12} and H_{12} show some variation with composition although this is only large for systems where there is a strong specific interaction between the components. There is a tendency of T_{12} and H_{12} at a certain composition to increase with the strength of interaction of the components, but this is not well-defined and T_{12} and H_{12} cannot generally be regarded as a measure of the strength of interaction [37].

A perusal of table 2 shows that the variations and signs of d_{12} are similar to those of $\Delta\eta$ and thereby supports our conclusion. It is also seen that, T_{12} and H_{12} values are positive for all binary mixtures and are almost identical and do not change appreciably with the change of composition of binary mixtures.

3.5. *Isentropic compressibility*

Table 5 contains the sound velocity (u), isentropic compressibility (K_S) and deviations in isentropic compressibility (ΔK_S) data for the binary mixtures and calculated at 298.15 K.

Isentropic compressibility K_S and deviations in isentropic compressibility ΔK_S were calculated from experimental densities (ρ) and speed of sound (u) using the following equations

$$K_S = (u^2\rho)^{-1} \tag{7}$$

$$\Delta K_S = K_S - \sum_{i=1}^{j}(x_i K_{S,i}) \tag{8}$$

where K_S gives the isentropic compressibility for the ith component of the mixture.

The ultrasonic speeds are given in table 3, together with the isentropic compressibility K_S and deviations in isentropic compressibility ΔK_S for 2-ethoxyethanol + 1-alkanol mixtures at 298.15 K. Experimental values for ΔK_S are plotted against mole fraction of 2-ethoxyethanol in figure 3.

We have attempted to explain the physico-chemical behavior of the mixtures in order to know the nature of molecular interactions between the components by various acoustical parameters calculated using the speed of sound and density data. Various parameters such as specific acoustic impedance Z, intermolecular free length L_f, van der Waal's constant b, molecular radius r, geometrical volume B, molar surface area Y, available volume V_a, molar speed of sound R, relative association R_A, collision factor S and molecular association M_A has been calculated using the following relations:

$$Z = u\rho \tag{9}$$

$$b = \left(\frac{M}{\rho}\right) - \left(\frac{RT}{\rho^2 u^2}\right)\left\{\left[1 + \left(\frac{Mu^2}{3RT}\right)\right]^{1/2} - 1\right\} \tag{10}$$

$$r = \left(\frac{3b}{16\pi N}\right)^{1/3} \tag{11}$$

$$L_f = \frac{K}{u\rho^{1/2}} \tag{12}$$

$$B = \frac{4}{3}\pi r^3 N \tag{13}$$

$$Y = (36\pi N B^2)^{1/3} \tag{14}$$

$$V_a = V - V_0 \tag{15}$$

$$R = \frac{Mu^{1/3}}{\rho} \tag{16}$$

$$S = \frac{uV}{u_\infty B} \tag{17}$$

Table 3. Values of ultrasonic speeds (u), isentropic compressibility (K_S), deviations in isentropic compressibility (ΔK_S) for binary mixtures of 2-ethoxyethanol + methanol, ethanol, propanol, butanol, pentanol, hexanol, heptanol and octanol at 298.15 K.

x_1	u (m s^{-1})	$K_S \times 10^{12}$ (Pa^{-1})	$\Delta K_S \times 10^{12}$ (Pa^{-1})	x_1	u (m s^{-1})	$K_S \times 10^{12}$ (Pa^{-1})	$\Delta K_S \times 10^{12}$ (Pa^{-1})
2-Ethoxyethanol + methanol					2-Ethoxyethanol + ethanol		
0	1104.2	1042.63	0	0	1142.0	975.89	0
0.0380	1130.9	981.86	−45.11	0.0538	1174.6	907.67	−49.66
0.0816	1151.5	931.90	−77.10	0.1133	1199.2	856.88	−79.88
0.1322	1167.5	886.36	−101.80	0.1798	1220.6	813.83	−100.01
0.1916	1188.6	842.09	−121.60	0.2542	1238.6	777.63	−110.49
0.2623	1205.1	803.78	−130.80	0.3383	1253.7	746.68	−112.40
0.3478	1220.5	771.62	−127.72	0.4341	1265.2	721.24	−104.80
0.4534	1233.1	744.02	−111.81	0.5440	1275.7	697.77	−90.30
0.5871	1246.9	716.60	−84.14	0.6716	1288.2	673.06	−70.96
0.7619	1266.2	684.13	−44.62	0.8215	1301.4	648.68	−43.60
1	1309.0	630.65	0	1	1309.0	630.65	0
2-Ethoxyethanol + propanol					2-Ethoxyethanol + butanol		
0	1209.4	806.49	0	0	1240.2	806.49	0
0.0690	1224.6	790.01	−17.36	0.0837	1245.0	790.01	−1.76
0.1429	1238.7	770.83	−30.71	0.1706	1252.3	770.83	−5.67
0.2223	1249.6	749.95	−37.90	0.2606	1261.4	749.95	−10.71
0.3078	1259.5	728.42	−41.50	0.3541	1271.4	728.42	−15.80
0.4001	1268.0	708.33	−41.43	0.4513	1280.6	708.33	−18.81
0.5001	1276.6	689.16	−39.01	0.5523	1289.3	689.16	−20.21
0.6088	1285.0	672.37	−33.70	0.6574	1296.1	672.37	−18.52
0.7273	1293.6	657.40	−26.11	0.7669	1301.2	657.40	−14.24
0.8572	1302.5	644.23	−15.88	0.8810	1304.8	644.23	−7.35
1	1309.0	630.65	0	1	1309.0	630.65	0
2-Ethoxyethanol + pentanol					2-Ethoxyethanol + hexanol		
0	1277.2	755.71	0	0	1328.3	695.34	0
0.0980	1281.8	741.34	−2.11	0.1119	1324.3	691.70	3.60
0.1965	1286.5	727.16	−3.98	0.2209	1320.9	687.23	6.18
0.2954	1291.0	713.23	−5.54	0.3270	1318.0	682.02	7.83
0.3947	1295.1	699.89	−6.46	0.4305	1315.7	676.16	8.67
0.4945	1298.8	687.10	−6.77	0.5314	1314.2	669.44	8.47
0.5947	1301.6	675.20	−6.14	0.6297	1313.1	662.24	7.64
0.6953	1303.8	663.86	−4.89	0.7257	1312.2	654.55	6.15
0.7964	1305.6	652.85	−3.26	0.8193	1311.5	646.56	4.22
0.8980	1307.1	641.99	−1.42	0.9108	1310.7	638.33	1.91
1	1309.0	630.65	0	1	1309.0	630.65	0
2-Ethoxyethanol + heptanol					2-Ethoxyethanol + octanol		
0	1331.5	687.28	0	0	1347.7	669.96	0
0.1253	1326.4	686.72	6.54	0.1384	1339.3	671.52	7.01
0.2438	1322.3	683.52	10.04	0.2654	1332.8	671.02	11.49
0.3559	1319.2	678.85	11.73	0.3825	1327.3	669.33	14.40
0.4623	1316.5	673.94	12.84	0.4907	1322.5	666.64	15.97
0.5632	1314.2	668.51	13.12	0.5910	1318.7	662.89	16.16
0.6592	1312.5	661.87	11.92	0.6843	1315.6	658.17	15.11
0.7505	1311.1	655.00	10.22	0.7713	1313.3	652.39	12.75
0.8376	1309.9	647.96	8.11	0.8525	1311.5	645.84	9.39
0.9207	1309.7	639.34	4.20	0.9286	1310.2	638.54	5.08
1	1309.0	630.65	0	1	1309.0	630.65	0

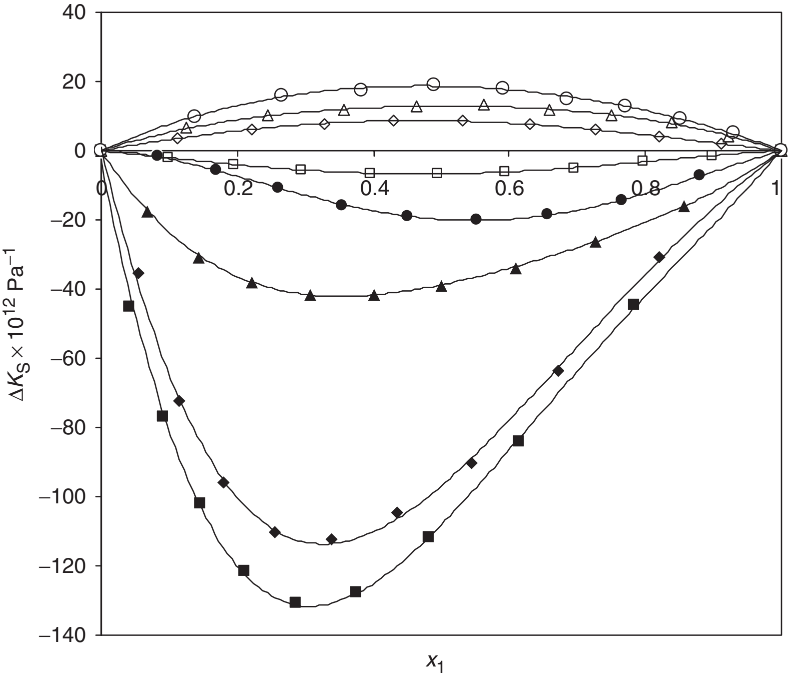

Figure 3. Deviations (ΔK_S) for binary mixtures of 2-ethoxyethanol with methanol (■), ethanol (◆), 1-propanol (▲), 1-butanol (●), 1-pentanol (□), 1-hexanol (◇), 1-heptanol (△) and 1-octanol (○) at 298.15 K.

$$M_A = \left[\left(\frac{u_{\text{mix}}}{\Sigma x_i u_i}\right)^2 - 1\right] \tag{18}$$

$$R_A = \left(\frac{L_{\text{f}}}{L_{\text{f}'}}\right)^3 \tag{19}$$

where K is a temperature-dependent constant, V_0 is volume at absolute zero, u_∞ is taken as 1600 m s^{-1}. These parameters are listed in table 4 for the pure components and in table 5 for the binary mixtures. Plots of L_{f}, Z, V_{a} and R against the mole fraction of 2-ethoxyethanol (x_1) are shown in figures 4–7, respectively.

It is observed that, the value of specific acoustic impedance Z, increases with increasing x_1 for all the mixtures, while the L_{f} behaves in an opposite manner. The graphs do not show any sudden variation in their behavior, thereby implying the absence of complex formation [16].

The R_{A} values increase with increasing x_1 for all the mixtures which signifies that the 1–2 interactions in these mixtures are not strongly dissociative. The decrease in L_{f} and V_{a} with increase in x_1 indicates significant interaction between the mixing molecules [28].

From figure 3, it is evident that the ΔK_S values are negative for lower monoalcohols but the magnitude of negative values diminishes and the positive values increases with

Table 4. van der Waal's constant b, molecular radius r, geometrical volume B, collision factor S, molar speed of sound R, available volume V_a, intermolecular free length L_f, molar volume at absolute zero V_0, molar surface area Y and specific acoustic impedance Z of the pure components at 298.15 K.

Component	$b \times 10^5$ (m^3)	r (nm)	$B \times 10^5$ (m^3 mol^{-1})	S	$R \times 10^6$ [m^{3}mol^{-1} (m s^{-1})$^{1/3}$]	$V_a \times 10^5$ (m^3)	L_f (Å)	$V_0 \times 10^5$ (m^3)	$Y \times 10^{-4}$ (Å)	$Z \times 10^{-3}$ (Kg m^2 s^{-1})
2-Ethoxyethanol	9.17	0.209	2.29	3.48	1065.30	1.77	0.5165	7.97	32.94	1211.35
Methanol	3.69	0.154	0.92	3.05	420.98	1.26	0.6641	2.81	17.95	868.61
Ethanol	5.38	0.174	1.34	0.92	613.01	1.67	0.6425	4.19	23.08	897.29
Propan-1-ol	6.97	0.190	1.74	3.26	801.07	1.84	0.6015	5.68	27.45	966.71
Butan-1-ol	8.59	0.204	2.15	3.32	987.77	2.07	0.5841	7.13	31.54	999.79
Pentan-1-ol	10.21	0.216	2.55	3.40	1179.00	2.19	0.5654	8.67	35.40	1036.06
Hexan-1-ol	11.84	0.227	2.96	3.52	1378.00	2.13	0.5424	10.41	39.08	1082.70
Heptan-1-ol	13.42	0.237	3.35	3.51	1557.70	2.38	0.5392	11.78	42.47	1092.76
Octan-1-ol	15.06	0.246	3.77	3.54	1750.42	2.50	0.5323	13.351	45.87	1107.54

Table 5. Speeds of sound, isentropic compressibility and excess isentropic compressibility, deviation of speed of sound, excess intermolecular free length and excess acoustic impedance of alkan-1-ol + 2-ethoxyethanol at 298.15 K.

x_1	K_S (TPa^{-1})	L_f (Å)	$R \times 10^6$ [m^3 mol^{-1} (m s^{-1})$^{1/3}$]	R_A	M_A	$V_a \times 10^5$ (m^3)	$Z \times 10^{-3}$
2-Ethoxyethanol + methanol							
0.0380	977.74	0.6431	370.86	0.9073	−0.4828	1.20	904.38
0.0816	927.53	0.6264	384.99	0.9170	−0.4738	1.17	936.28
0.1322	887.44	0.6127	402.39	0.9281	−0.4660	1.15	965.17
0.1916	842.05	0.5969	424.43	0.9381	−0.4596	1.12	999.14
0.2623	805.36	0.5837	451.84	0.9497	−0.4548	1.11	1030.36
0.3478	772.16	0.5715	488.29	0.9617	−0.4529	1.13	1061.10
0.4534	744.05	0.5610	539.29	0.9744	−0.4548	1.18	1089.94
0.5871	716.16	0.5504	617.87	0.9864	−0.4620	1.27	1119.84
0.7619	684.14	0.5380	756.05	0.9962	−0.4766	1.44	1154.39
2-Ethoxyethanol + ethanol							
0.0538	907.71	0.6197	539.56	0.8946	−0.5187	1.58	937.92
0.1133	856.90	0.6021	560.05	0.9029	−0.5114	1.51	973.15
0.1798	813.87	0.5868	583.40	0.9122	−0.5049	1.45	1006.63
0.2542	777.66	0.5736	610.62	0.9226	−0.4992	1.42	1038.19
0.3383	746.66	0.5620	643.28	0.9341	−0.4945	1.40	1068.28
0.4341	721.21	0.5524	683.56	0.9467	−0.4913	1.41	1095.92
0.5440	697.79	0.5433	735.61	0.9598	−0.4898	1.45	1123.38
0.6716	673.08	0.5336	806.51	0.9727	−0.4904	1.49	1153.33
0.8215	648.63	0.5238	908.28	0.9856	−0.4936	1.58	1184.66
2-Ethoxyethanol + propanol							
0.0690	822.43	0.5899	711.17	0.8958	−0.5556	1.78	992.91
0.1429	792.47	0.5790	732.34	0.9052	−0.5483	1.73	1018.71
0.2223	767.51	0.5698	755.46	0.9157	−0.541	1.70	1042.67
0.3078	744.60	0.5613	781.48	0.9267	−0.5339	1.68	1066.29
0.4001	723.97	0.5534	811.03	0.9383	−0.5271	1.68	1089.34
0.5001	703.92	0.5457	845.34	0.9499	−0.5208	1.67	1112.81
0.6088	684.77	0.5382	885.67	0.9616	−0.5151	1.68	1136.45
0.7273	665.77	0.5307	933.91	0.9738	−0.5097	1.69	1161.12
0.8572	646.86	0.5231	992.71	0.9863	−0.5047	1.72	1186.90
2-Ethoxyethanol + butanol							
0.0837	790.05	0.5781	874.65	0.8973	−0.5705	2.04	1016.67
0.1706	770.86	0.5711	890.28	0.9072	−0.5634	2.01	1035.90
0.2606	749.89	0.5632	907.40	0.9169	−0.5562	1.96	1057.18
0.3541	728.41	0.5551	925.91	0.9267	−0.5489	1.91	1079.80
0.4513	708.30	0.5474	945.54	0.9371	−0.5414	1.87	1102.47
0.5523	689.17	0.5400	966.52	0.9480	−0.5336	1.83	1125.43
0.6574	672.33	0.5333	988.67	0.9599	−0.5256	1.80	1147.57
0.7669	657.42	0.5274	1012.20	0.9728	−0.5172	1.78	1169.00
0.8810	644.26	0.5221	1037.50	0.9863	−0.5088	1.78	1189.59
2-Ethoxyethanol + pentanol							
0.0980	741.34	0.5600	1036.97	0.8934	−0.5888	2.14	1052.36
0.1965	727.16	0.5546	1040.48	0.9031	−0.5809	2.09	1068.95
0.2954	713.26	0.5493	1043.95	0.9132	−0.5725	2.04	1085.99
0.3947	699.93	0.5442	1047.35	0.9237	−0.5638	1.99	1103.17
0.4945	687.08	0.5391	1050.66	0.9348	−0.5547	1.94	1120.60
0.5947	675.20	0.5345	1053.74	0.9465	−0.5451	1.91	1137.86
0.6953	663.89	0.5300	1056.69	0.9588	−0.5349	1.87	1155.30
0.7964	652.85	0.5255	1059.56	0.9719	−0.5241	1.84	1173.21
0.8980	641.99	0.5212	1062.36	0.9857	−0.5125	1.81	1191.68

(*Continued*)

Table 5. Continued.

x_1	K_S (TPa^{-1})	L_f (Å)	$R \times 10^6$ [m^3 mol^{-1} (m s^{-1})$^{1/3}$]	R_A	M_A	$V_a \times 10^5$ (m^3)	$Z \times 10^{-3}$
2-Ethoxyethanol + hexanol							
0.1119	691.65	0.5409	1194.66	0.8874	−0.6135	1.20	1091.75
0.2209	687.22	0.5392	1176.73	0.8985	−0.6032	1.17	1101.63
0.3270	682.07	0.5372	1159.86	0.9099	−0.5924	1.15	1112.39
0.4305	676.20	0.5349	1144.01	0.9216	−0.5811	1.12	1124.00
0.5314	669.44	0.5322	1129.16	0.9334	−0.5694	1.11	1136.65
0.6297	662.22	0.5293	1115.14	0.9454	−0.5572	1.13	1150.01
0.7257	654.53	0.5262	1101.84	0.9580	−0.5442	1.18	1164.32
0.8193	646.56	0.5230	1089.20	0.9711	−0.5306	1.27	1179.30
0.9108	638.33	0.5197	1077.11	0.9850	−0.5160	1.44	1195.23
2-Ethoxyethanol + heptanol							
0.1253	686.72	0.5390	1331.37	0.8905	−0.6119	2.32	1097.86
0.2438	683.55	0.5378	1287.40	0.9011	−0.6019	2.25	1106.37
0.3559	678.90	0.5359	1248.55	0.9123	−0.5910	2.18	1116.57
0.4623	673.96	0.5340	1213.83	0.9233	−0.5801	2.12	1127.06
0.5632	668.51	0.5318	1182.65	0.9347	−0.5687	2.06	1138.23
0.6592	661.84	0.5291	1154.56	0.9470	−0.5562	1.99	1151.19
0.7505	654.96	0.5264	1129.09	0.9593	−0.5435	1.94	1164.52
0.8376	647.92	0.5236	1105.85	0.9718	−0.5304	1.89	1178.26
0.9207	639.38	0.5201	1084.81	0.9851	−0.5161	1.83	1194.18
2-Ethoxyethanol + octanol							
0.1384	671.52	0.5330	1461.25	0.8903	−0.6175	2.41	1109.65
0.2654	671.06	0.5328	1385.10	0.9011	−0.6066	2.32	1114.39
0.3825	669.29	0.5321	1321.61	0.9122	−0.5951	2.24	1122.97
0.4907	666.69	0.5311	1267.83	0.9236	−0.5834	2.16	1131.60
0.5910	662.89	0.5296	1221.83	0.9351	−0.5712	2.09	1142.32
0.6843	658.20	0.5277	1182.02	0.9470	−0.5586	2.02	1154.83
0.7713	652.40	0.5254	1147.29	0.9593	−0.5452	1.96	1167.13
0.8525	645.84	0.5227	1116.70	0.9722	−0.5312	1.89	1180.61
0.9286	638.54	0.5197	1089.59	0.9855	−0.5163	1.83	1195.30

the increasing chain length of the alcohols. The values of ΔK_S in terms of negativity are enhanced by the following order:

$$\text{Methanol} > \text{Ethanol} > \text{1-Propanol} > \text{1-Butanol} > \text{1-Pentanol}$$

$$> \text{1-Hexanol} > \text{1-Heptanol} > \text{1-Octanol}$$

These results can be explained in terms of molecular interactions and structural effects. Positive ΔK_S values are due to the breaking of interactions and the corresponding disruption of molecular order in the pure components [42]. The donor–acceptor interaction between the 2-ethoxyethanol and the alcohols play an important part for the mixtures containing lower alcohols like methanol, ethanol, 1-propanol where there is strong specific interaction between the component molecules leading to negative value of ΔK_S. Interactions between the molecules of 2-ethoxyethanol or monoalcohols are broken in the mixing process; the breaking leads to positive K_S^E values for the mixture containing higher chain length of alcohols as compared to the lower alcohols. There is a parallel in the qualitative behavior of ΔK_S and V^E curves.

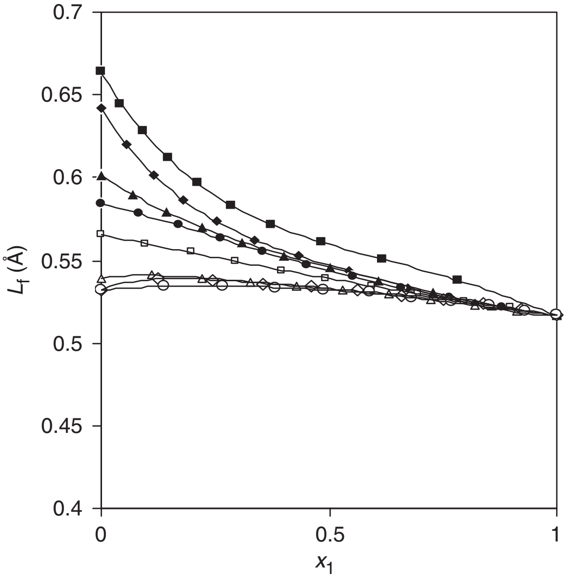

Figure 4. L_f values for the experimental mixtures against the mole fraction of 2-ethoxyethanol, methanol (■), ethanol (◆), 1-propanol (▲), 1-butanol (●), 1-pentanol (□), 1-hexanol (◇), 1-heptanol (Δ) and 1-octanol (○) at 298.15 K.

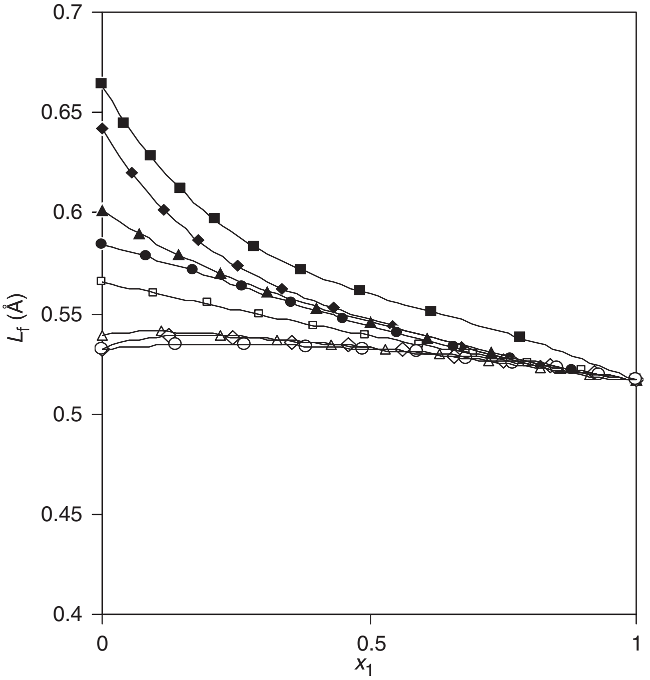

Figure 5. Z values for the experimental mixtures against the mole fraction of 2-ethoxyethanol, methanol (■), ethanol (◆), 1-propanol (▲), 1-butanol (●), 1-pentanol (□), 1-hexanol (◇), 1-heptanol (Δ) and 1-octanol (○) at 298.15 K.

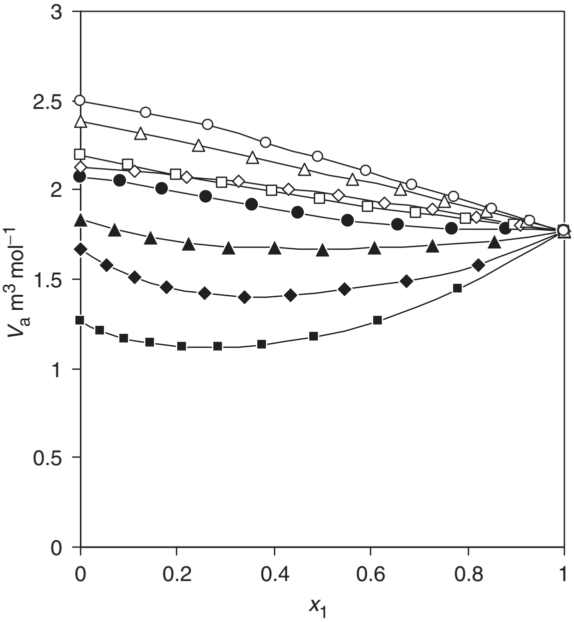

Figure 6. V_a values for the experimental mixtures against the mole fraction of 2-ethoxyethanol, methanol (■), ethanol (◆), 1-propanol (▲), 1-butanol (●), 1-pentanol (□), 1-hexanol (◇), 1-heptanol (Δ) and 1-octanol (○) at 298.15 K.

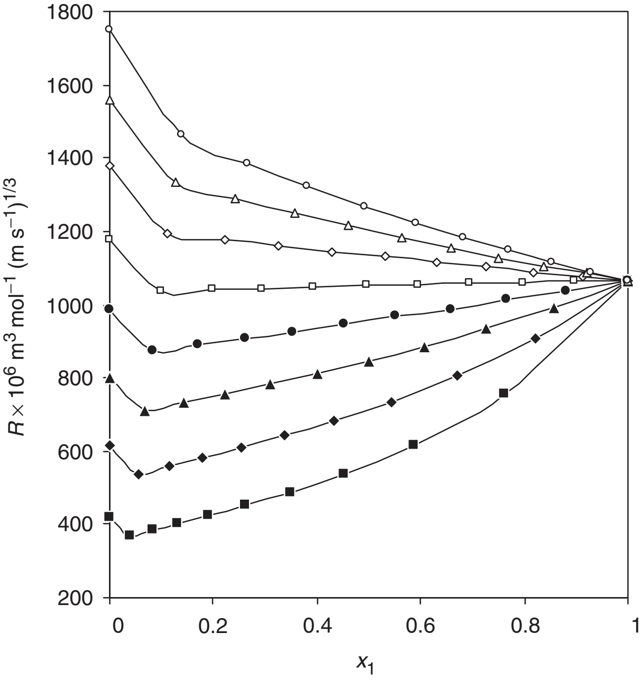

Figure 7. R values for the experimental mixtures against the mole fraction of 2-ethoxyethanol, methanol (■), ethanol (◆), 1-propanol (▲), 1-butanol (●), 1-pentanol (□), 1-hexanol (◇), 1-heptanol (Δ) and 1-octanol (○) at 298.15 K.

3.6. *Redlich–Kister polynomial equation*

The excess properties (V^{E}, $\Delta\eta$, $\Delta G^{*\mathrm{E}}$ and ΔK_{S}) were fitted to the Redlich–Kister polynomial equation [43],

$$Y^{\mathrm{E}} = x_1 x_2 \sum_{i=1}^{K} A_i (x_1 - x_2)^i \tag{20}$$

where Y^{E} refers to excess properties, x_1 is the mole fraction of 2-ethoxyethanol and x_2 is that of the other component of the binary mixtures, respectively. The coefficients (A_i) were obtained by fitting equation (20) to experimental results using a least squares regression method. In each case, the optimal number of coefficients was ascertained from an approximation of the variation in the standard deviation (σ). The calculated values of A_i along with the tabulated standard deviations (σ) are listed in table 6. The standard deviation (σ) was calculated using the equation,

$$\sigma^2 = \frac{1}{n - m} \left(Y^{\mathrm{E}}_{\mathrm{exp}} - Y^{\mathrm{E}}_{\mathrm{cal}} \right)^2 \tag{21}$$

where n is the number of data points and m is the number of coefficients.

3.7. *Predictions of sound speeds*

The sound speeds of binary mixtures are often predicted by free length theory (FLT), collision factor theory (CFT), the Nomoto equation, the Vandeal Vangael (VV) ideal mixing relation, the impedance dependence (ID) relation, etc. For comparison, the theoretical values of the sound speed u has been calculated by using the above five theories and empirical equations. The following final relations were used for calculating sound speeds.

According to FLT [12], the speed of sound is given by,

$$u_{\mathrm{FLT}} = \frac{K_{\mathrm{S}}}{L_{\mathrm{f}} \rho^{1/2}} \tag{22}$$

The free length L_{f} is obtained by:

$$L_{\mathrm{f}} = \frac{V - \sum_i x_i V_{0i}}{\sum_i x_i y_i} \tag{23}$$

where, V_{0i} is the molar volume of the pure component i at absolute zero and is given by Sugden's formula,

$$V_{0i} = V_i \left(\frac{1 - T}{T_{\mathrm{c}i}} \right)^{0.3} \tag{24}$$

where T_{c} is the critical temperature for the pure components.

Y_i is the surface area per mole for the pure component i and is given by,

$$Y_i = \left(36\pi N V_{0i}^2 \right)^{1/3} \tag{25}$$

Collision factor theory [15],

$$u_{\mathrm{CFT}} = u_{\infty}[x_1 S_1] + \left[\frac{x_1 B_1 + x_2 B_2}{V_{12}} \right] \tag{26}$$

Nomoto equation [56],

$$u_N = \left[\frac{x_1 R_1 + x_2 R_2}{x_1 V_1 + x_2 V_2}\right]^3 \tag{27}$$

Vandeal Vangael [57] ideal mixing relation

$$\frac{1}{x_1 M_1 + x_2 M_2} - \frac{1}{u_{\text{mix}}^2} = \frac{x_1}{M_1 u_1^2} + \frac{x_2}{M_2 u_2^2} \tag{28}$$

Table 6. Redlich–Kister coefficients and standard deviations (σ) for the binary mixtures of 2-ethoxyethanol + methanol, ethanol, 1-propanol, 1-butanol, 1-pentanol, 1-hexanol, 1-heptanol and 1-octanol at $T = 298.15\,\text{K}$.

Excess property	A_0	A_1	A_2	A_3	A_4	σ
2-Ethoxyethanol + methanol						
$V^E \times 10^6$ (m^3 mol^{-1})	3.16995	−0.19613	9.25258	–	–	0.49292
$\Delta\eta$ (mPa S)	0.45326	−0.06186	0.15702	–	–	0.00207
ΔG^{*E} (J mol^{-1})	3479.34	−1432.39	1298.143	−2197.38	−1306.92	2.65174
$\Delta K_S \times 10^{12}$ (Pa^{-1})	−410.03	418.6652	−260.832	−231.895	186.4851	0.26013
2-Ethoxyethanol + ethanol						
$V^E \times 10^6$ (m^3 mol^{-1})	−0.7623	0.1832	−0.14364	−0.26987	0.10883	0.0013
$\Delta\eta$ (mPa s)	0.31223	0.00246	–	–	–	0.00039
ΔG^{*E} (J mol^{-1})	1146.97	−231.093	71.5142	–	–	0.69754
$\Delta K_S \times 10^{12}$ (Pa^{-1})	−384.08	270.8331	−468.176	−422.724	1819.404	0.0000
2-Ethoxyethanol + 1-propanol						
$V^E \times 10^6$ (m^3 mol^{-1})	−0.4525	0.63921	0.95591	−4.03612	−7.6092	0.0000
$\Delta\eta$ (mPa s)	−0.4924	0.31418	−0.06658	0.06129	–	0.00028
ΔG^{*E} (J mol^{-1})	−576.47	417.0622	−40.3791	4.6357	−17.0632	0.29871
$\Delta K_S \times 10^{12}$ (Pa^{-1})	−155.20	77.52434	−64.4247	9.7579	–	0.2561
2-Ethoxyethanol + 1-butanol						
$V^E \times 10^6$ (m^3 mol^{-1})	0.35678	0.0052	−0.31737	–	–	0.00057
$\Delta\eta$ (mPa s)	−0.7822	0.34872	0.3328	–	–	0.00137
ΔG^{*E} (J mol^{-1})	−778.52	278.1309	472.5683	71.41829	–	1.65605
$\Delta K_S \times 10^{12}$ (Pa^{-1})	−79.574	−24.6251	50.09514	−3.56549	–	0.13396
2-Ethoxyethanol + 1-pentanol						
$V^E \times 10^6$ (m^3 mol^{-1})	0.85714	0.30517	−0.11772	–	–	0.00185
$\Delta\eta$ (mPa s)	−1.6096	0.90572	0.24544	−0.11007	0.1719	0.00167
ΔG^{*E} (J mol^{-1})	−794.99	56.02497	511.5964	241.3453	–	1.67093
$\Delta K_S \times 10^{12}$ (Pa^{-1})	−27.041	4.06096	24.65183	6.48988	−105.956	0.0000
2-Ethoxyethanol + 1-hexanol						
$V^E \times 10^6$ (m^3 mol^{-1})	0.94412	0.36152	0.52195	0.15616	−0.25529	0.00331
$\Delta\eta$ (mPa s)	−2.4006	1.13292	0.20876	0.37026	–	0.0073
ΔG^{*E} (J mol^{-1})	−958.04	416.3159	634.2876	439.3159	–	5.67164
$\Delta K_S \times 10^{12}$ (Pa^{-1})	34.5516	−5.80756	−7.03454	4.42241	7.98105	0.03121
2-Ethoxyethanol + 1-heptanol						
$V^E \times 10^6$ (m^3 mol^{-1})	1.58018	0.71001	0.21536	–	–	0.00249
$\Delta\eta$ (mPa s)	−3.1936	1.24977	0.13886	–	–	0.01012
ΔG^{*E} (J mol^{-1})	−436.66	432.5375	−307.039	−728.232	1304.31	2.29942
$\Delta K_S \times 10^{12}$ (Pa^{-1})	51.6636	3.55552	12.76334	−7.78112	–	0.19454
2-Ethoxyethanol + 1-octanol						
$V^E \times 10^6$ (m^3 mol^{-1})	1.65059	0.51687	0.46603	0.26796	–	0.00234
$\Delta\eta$ (mPa s)	−4.0219	0.70295	0.44447	0.61789	–	0.00719
ΔG^{*E} (J mol^{-1})	74.1220	−213.353	649.6937	2647.439	−2950.2	0.0000
$\Delta K_S \times 10^{12}$ (Pa^{-1})	64.1492	14.7396	3.89818	−6.8375	2.27945	0.0169

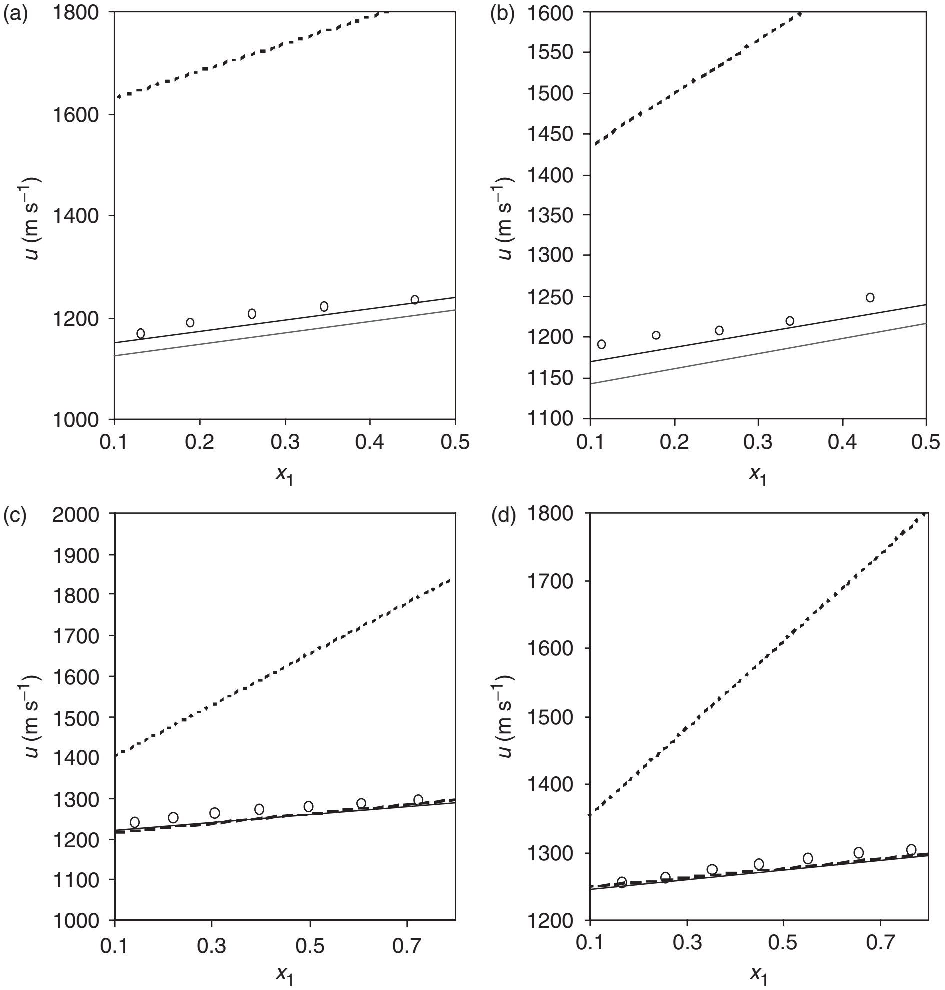

Figure 8(a–h). Experimental and calculated ultrasonic speed u of [2-ethoxyethanol] + [methanol (a), ethanol (b), 1-propanol (c), 1-butanol (d), 1-pentanol (e), 1-hexanol (f), 1-heptanol (g) and 1-octanol (h)] at 298.15 K. [$u_{\text{experimental}}$ (○○○○○); u_{FLT} (-------); u_{Nomoto} (– – – –); u_{CFT} (·········).].

Impedance dependence relation [58,59]

$$u = \frac{\sum x_i Z_i}{x_i \rho_i} \tag{29}$$

where, K_S, S, B, R, Z, ρ are the isentropic compressibility, collision factor, geometrical volume, molar speed of sound, specific acoustic impedance and density respectively for pure 2-ethoxyethanol (**1**), 1-alkanols (**2**), and the mixtures (12), u_∞ is a constant value taken as 1600 m s^{-1}.

The compositional dependence of the experimental and sound speeds calculated by the free length theory (FLT), collision factor theory (CFT) and the Nomoto equation of the binary mixtures of 2-ethoxyethanol and 1-alkanols are shown in figure 8(a–h). The results show that the Nomoto equation and the collision factor theory predicts the

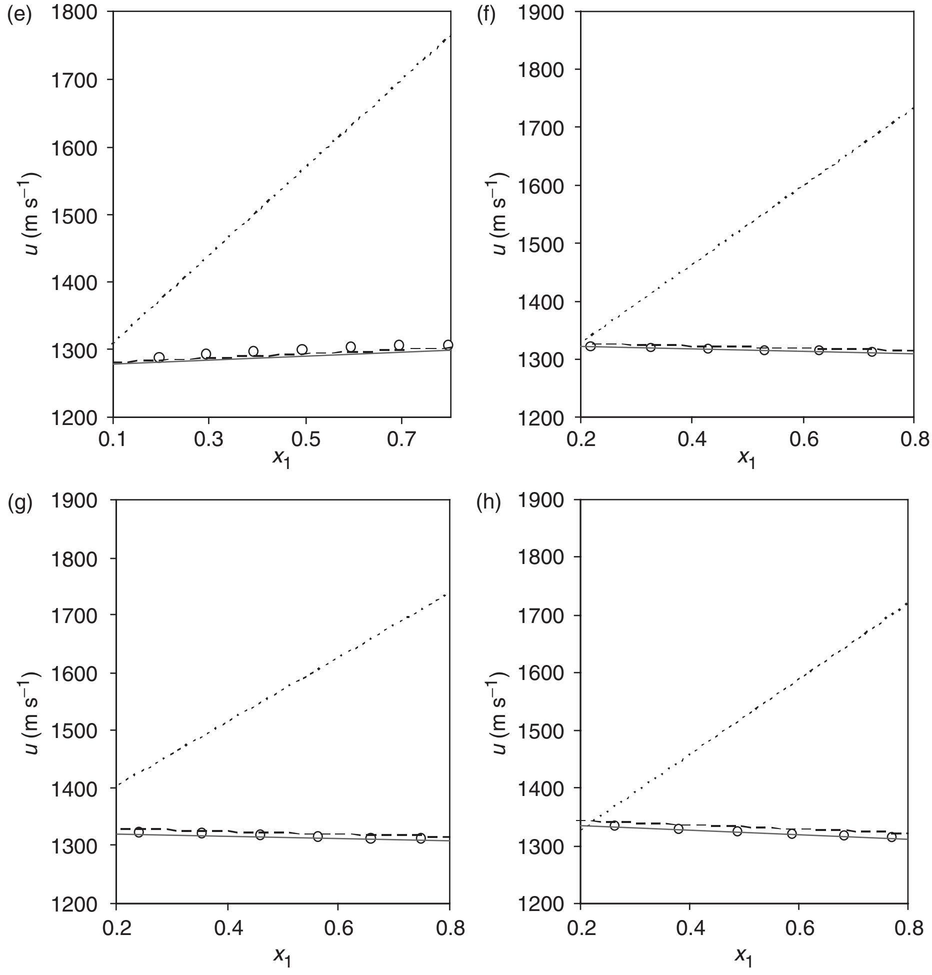

Figure 8(a–h). Continued.

experimental data extremely well, whereas the free length theory gives the maximum deviation for the present set of binary mixtures. Table 7 summarizes the deviations for different prediction methods. Based on deviation values obtained, the following order for the relative predictive capability for each of the methods is obtained,

$$\text{Nomoto} \geq \text{CFT} > \text{ID} \geq \text{VV} > \text{FLT}$$

4. Conclusions

In this work, eight binary mixtures have been studied in terms of excess molar volumes, viscosity deviations, acoustic impedance, intermolecular free length, isentropic

Table 7. Values of standard deviation (%) for sound speeds by various methods.

Solvent mixture	σ (%)				
	u_{CFT}	u_{FLT}	u_{Nomoto}	U_{VV}	u_{ID}
2-Ethoxyethanol + methanol	0.11	0.85	0.10	0.63	0.15
2-Ethoxyethanol + ethanol	0.20	1.19	0.12	0.35	0.16
2-Ethoxyethanol + 1-propanol	0.04	1.35	0.05	0.13	0.05
2-Ethoxyethanol + 1-butanol	0.05	1.44	0.04	0.07	0.04
2-Ethoxyethanol + 1-pentanol	0.02	1.59	0.02	0.02	0.02
2-Ethoxyethanol + 1-hexanol	0.01	1.79	0.02	0.01	0.01
2-Ethoxyethanol + 1-heptanol	0.01	1.71	0.02	0.02	0.01
2-Ethoxyethanol + 1-octanol	0.01	1.76	0.04	0.02	0.02

compressibility and interaction parameters. The investigation was carried out to explore the nature of interactions between the mixing components. It may be concluded from our effort that, the strength of associative interaction between the unlike molecules weakens with the increase of chain length of the 1-alkanols.

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