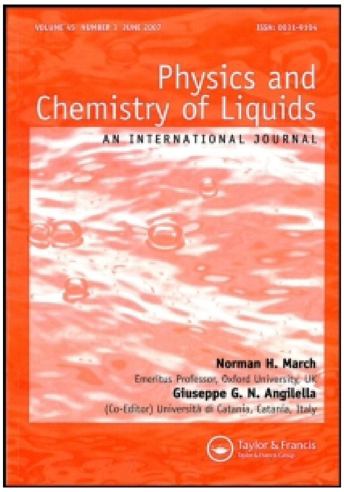

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Densities, viscosities, speeds of sound and refractive indices of binary mixtures of tetrahydrofuran with 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol at 298.15, 303.15, 308.15 and 313.15 K

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Densities, viscosities, speeds of sound and refractive indices of binary mixtures of tetrahydrofuran with 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol at 298.15, 303.15, 308.15 and 313.15 K

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Densities (ρ), dynamic viscosities (η), refractive indices (n_D) and speeds of sound (u) of binary mixtures of tetrahydrofuran with 1-hexanol, 1-heptanol, 1-octanol, 1-nonanol and 1-decanol over the entire range of mole fraction, at temperatures 298.15, 303.15, 308.15 and 313.15 K and atmospheric pressure have been measured. Excess functions such as excess molar volume (V^E), deviations in molar refractivity (ΔR_m), deviations in speed of sound (u^D), excess isentropic compressibilities (k_s^E) and viscosity deviations ($\Delta\eta$) have been calculated and fitted to the Redlich–Kister-type polynomial equation. The viscosity data were correlated with semi-empirical equations. The speeds of sound have been analysed in terms of collision factor theory and free length theory. The behaviour of V^E, ΔR_m and u^D over the entire range of mole fraction, exhibited positive deviations, while k_s^E and $\Delta\eta$ showed negative variations.

Keywords: densities; viscosities; speed of sound; refractive indices; excess molar volume; tetrahydrofuran; 1-alkanols

1. Introduction

The knowledge of transport and thermodynamic properties of binary liquid mixtures containing cyclic ethers are of considerable interest from the viewpoint of academic and many industrial processes [1–4]. Cyclic ethers are excellent solvents of polymers and are used in polymer synthesis [5]. Tetrahydrofuran (THF) is often applied for hydroborations, used to synthesise primary alcohols, as well as for preparation of Grignard reagents, the most useful and versatile reagents known to the organic chemists; besides it is used together with alkan-1-ol in production of mixed solvents. Continuing our studies on the behaviour of ethers with hydrocarbons [6–9], we present here a systematic investigations involving densities (ρ), dynamic viscosities (η), speeds of sound (u) and refractive indices (n_D) for pure components and binary mixtures of THF + 1-hexanol, or 1-heptanol, or 1-octanol, or 1-nonanol, or 1-decanol over the whole composition range at 298.15, 303.15, 308.15 and 313.15 K and atmospheric pressure. From the measured values of the aforementioned physical properties, the corresponding magnitudes of excess molar volume (V^E), deviations in speed of sound (u^D), excess isentropic compressibilities (k_s^E), viscosity deviations ($\Delta\eta$) and deviations in the molar refractivity (ΔR_m) were derived.

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The excess properties data were fitted to the Redlich and Kister-type polynomial equation [10] to calculate the binary coefficients and estimate the SDs between experimental and predicted data. The dynamic viscosity data were correlated with equations of the three-body and four-body McAllister [11]. The speeds of sound in the binary mixtures have been predicted using Schaaffs' collision factor theory (CFT) [12] and Jacobson's intermolecular free length theory (FLT) [13] and compared with the experimental values. The variations of the excess functions with the molecular size of 1-alkanols at different temperatures were investigated. The refractive index data were correlated by Lorentz–Lorenz and Gladstone–Dale equations, reported elsewhere [14,15]. Although the physical properties data of the pure components, THF and 1-alkanols, are available in the literature [6,16–22], to the best of our knowledge no experimental data are available for these mixtures at the comparable conditions of this study.

2. Experimental section

2.1. *Chemicals*

The suppliers, purities, densities, dynamic viscosities, speeds of sound and refractive indices of chemicals used in this study are presented in Table 1.

All chemicals were stored in dark bottles over freshly activated molecular sieve (Union Carbide type 4A 1/16 inch, pellets) to minimise adsorption of moisture. The water content of the chemicals was checked by a Mettler Toledo DL39-KF coulometer, and it was found to be <0.0003 mass fraction.

The purity of the solvents was ascertained by comparing the measured densities, dynamic viscosities and speeds of sound of the pure components at 298.15 K with the available literature, as shown in Table 1. The reported experimental values of ρ, η and u conform closely to their corresponding literature values, with an average of the absolute value of deviation $3.6 \times 10^{-3}\,\mathrm{kg\,m^{-3}}$, $3.3 \times 10^{-3}\,\mathrm{mPa\,s}$ and $0.26\,\mathrm{m\,s^{-1}}$, respectively.

Table 1. Pure component properties and their comparison with the literature values at $T = 298.15$ K.

Compound	Supplier	ρ (kg m^{-3})	η (mPa s)	u (m s^{-1})	n_D
THF	Fluka	882.322 881.98 [16]	0.461 0.467 [16]	1278.93	1.4053 1.4049 [17]
1-Hexanol	Fluka	815.230 815.23 [6] 815.22 [18]	4.593 4.593 [19]	1304.72 1303.3 [20]	1.4154 1.4161 [17]
1-Heptanol	Fluka	818.732 818.7 [19]	5.944 5.942 [19]	1327.32	1.4224 1.4227 [21]
1-Octanol	Fluka	821.722 821.79 [22] 821.81 [18]	7.661 7.663 [19] 7.5981 [6]	1347.82 1347.43 [22]	1.4274 1.4276 [17]
1-Nonanol	Fluka	824.478 824.4 [19]	9.715 9.715 [19] 9.6921 [6]	1365.48	1.4318 1.4319 [21]
1-Decanol	Fluka	826.762 826.7 [17]	11.825 11.829 [19]	1380.01 1380.2 [20]	1.4356 1.4358 [21]

2.2. *Measurements*

Each sample mixture was prepared, on mass basis, by mixing the calculated volumes of liquid components in specially designed glass stoppered bottles. The mass measurements were carried out using an electronic balance (Mettler AT460) with a stated precision of $\pm 10^{-4}$ g. Triplicate measurements for mole fractions were averaged, and the uncertainty was found to be within $\pm 2 \times 10^{-4}$. Three samples were prepared for each system, and their densities, speeds of sound, viscosities and refractive indices were measured on the same day.

Densities and speeds of sound were measured at temperature intervals between 298.15 and 313.15 K using a tube vibrating densimeter and sound analyser, Anton-Paar DSA 5000. The details of measurements and calibration of the instrument were described in our earlier work [23]. For all mixtures and pure components, a triplicate of measurements was performed and the results were averaged. Respectively, the absolute values of deviations in measured ρ and u were $3 \times 10^{-3}\,\mathrm{kg\,m^{-3}}$ and $0.12\,\mathrm{m\,s^{-1}}$, while the uncertainties were within $\pm 6 \times 10^{-3}\,\mathrm{kg\,m^{-3}}$ and $\pm 0.13\,\mathrm{m\,s^{-1}}$.

Dynamic viscosity, η, measurements of all pure components and their binary mixtures were determined using a digital Anton Paar Stabinger Viscometer (model SVM 3000/G2). This instrument is made up of two measuring cells, one designed for measuring density of the samples based on the relation between oscillation period and density, while the other is used for dynamic viscosity measurements and consists of a tube filled with sample liquid rotating at a constant speed ω_1. A hollow measuring rotor (titanium rotor) of low density swims in this tube and is centred in a heavier sample liquid by buoyancy forces. The measuring gap is formed between the tube and the rotor. The rotor is guided axially by a built-in permanent magnet. The rotating magnetic field delivers the speed signal and induces eddy currents in the surrounding copper casing. The rotor speed ω_2 adopts an equilibrium between the viscosity-dependent driving torque, which is proportional to the speed difference between the outer tube and the inner rotor, and the retarding torque caused by eddy currents, which is proportional to the inner rotor speed.

The unambiguous measurement for the dynamic viscosity is calculated by the instrument according to the following expression:

$$\eta = \frac{k\omega_2}{\omega_1 - \omega_2}, \tag{1}$$

where k is the mean adjustment coefficient of the instrument. By the aid of built-in program and four certified standard samples supplied by the manufacturer, SHL109, SHM109, SCH100 and SHH110, of dynamic viscosities 3.058, 32.62, 99.51 and 1131 mPa s, respectively at 293.15 K, the whole instrument measuring ranges were adjusted automatically. After completing the instrument adjustment, the calibration of the instrument was achieved by a triplicate measurement of the reference samples at the interested temperatures with reproducibility ±0.35% of the measuring range. The estimated uncertainty in dynamic viscosity measurements was within $\pm 3 \times 10^{-3}$ mPa s with 95% confidence interval of all measurements. The temperature control of the cells was achieved by a solid-state thermostat and two integrated Pt-100 measuring sensors of temperature reproducibility of $\pm 10^{-2}$ K.

Refractive indices were measured for the sodium D-line at temperatures 293.15 and 303.15 K, using a thermostatic digital refractometer (ABBE Mark II Model No. 10481, Cambridge Instruments Inc. USA) with a precision of 10^{-4}. The details of measurement and calibration of the instrument are reported in our previous work [8], and the uncertainty in refractive indices was estimated to be better than $\pm 1.3 \times 10^{-4}$.

3. Results and discussion

The excess molar volume (V^{E}) was calculated using the following equation:

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

where x, M and ρ are mole fraction, molar mass and density, respectively. The subscripts i and m represent the pure components and the mixture, respectively.

The excess isentropic compressibility ($k_{\mathrm{s}}^{\mathrm{E}}$) was obtained from the relation

$$k_{\mathrm{s}}^{\mathrm{E}} = k_{\mathrm{s}} - k_{\mathrm{s}}^{\mathrm{id}}, \tag{3}$$

where k_{s} is the calculated isentropic compressibility ($=1/u^2\rho$) and $k_{\mathrm{s}}^{\mathrm{id}}$ is the isentropic compressibility for an ideal mixture of the components, and is estimated using the following relation [22]:

$$k_{\mathrm{s}}^{\mathrm{id}} = \sum \phi_i \left[k_{\mathrm{s},i}^{\mathrm{o}} + T V_i^{\mathrm{o}} \left(\alpha_i^{\mathrm{o}} \right)^2 / C_{\mathrm{p},i}^{\mathrm{o}} \right] - T \left(\sum x_i V_i^{\mathrm{o}} \right) \left(\sum \phi_i \alpha_i^{\mathrm{o}} \right)^2 \Big/ \sum x_i C_{\mathrm{p},i}^{\mathrm{o}}, \tag{4}$$

where ϕ_i is the volume fraction of i in the mixture stated in terms of the unmixed components, T is the temperature, and $k_{\mathrm{s},i}^{\mathrm{o}}$, V_i^{o}, α_i^{o} and $C_{\mathrm{p},i}^{\mathrm{o}}$ are, respectively, the isentropic compressibility, molar volume, isobaric expansion coefficient and molar isobaric heat capacity for pure component i. The values of $C_{\mathrm{p},i}^{\mathrm{o}}$ were taken from the DIPPR database [17] and are listed in Table 2.

The experimental data on density (ρ) for each of the pure liquids in the working temperature range, were fitted to the following equation [24]:

$$\rho = \sum_{i=1}^{3} A_i \left(\frac{T}{K} - 273.15 \right)^{i-1}, \tag{5}$$

where the coefficients A_i are given in Table 3. The values of α_i^{o}, defined as $\alpha_i^{\mathrm{o}} = V^{-1} \times (\mathrm{d}V/\mathrm{d}T)_{\mathrm{P}} = -\rho^{1}\ (\mathrm{d}\rho/\mathrm{d}T)_{\mathrm{p}}$, were calculated at each temperature using coefficients of Equation (5), and are shown in Table 4.

Table 2. The values of critical temperature and heat capacity of the pure components at $T=$ 298.15–313.15 K.

		$C_{\mathrm{p},i}^{\mathrm{o}}$ ($\mathrm{J\,mol^{-1}\,K^{-1}}$)			
	T_{c} (K)	$T=298.15$ K	$T=303.15$ K	$T=308.15$ K	$T=313.15$ K
THF	540.15	123.97	125.68	126.62	127.98
1-Hexanol	610.40	241.64	246.52	251.47	256.48
1-Heptanol	632.60	278.24	284.37	289.83	295.35
1-Octanol	652.50	308.39	313.60	320.12	325.98
1-Nonanol	670.70	337.33	342.40	348.18	354.40
1-Decanol	687.30	373.10	380.40	385.40	391.50

The deviations of the measured speeds of sound from their values in an ideal mixture were calculated from the following equations [25]:

$$u^{\mathrm{D}} = u - u^{\mathrm{id}}, \tag{6}$$

$$u^{\mathrm{id}} = V_{\mathrm{m}}^{\mathrm{id}}\left[\left(\sum x_i M_i\right) k_{\mathrm{s}}^{\mathrm{id}}\right]^{-0.5}, \tag{7}$$

where u^{id}, $V_{\mathrm{m}}^{\mathrm{id}}$, $k_{\mathrm{s}}^{\mathrm{id}}$, x_i and M_i are, respectively, the calculated speed of sound, molar volume, isentropic compressibility of the ideal solution, mole fraction and molar mass of the pure component.

Deviations in dynamic viscosity ($\Delta\eta$) were calculated using the following relation:

$$\Delta\eta = \eta_{\mathrm{m}} - \sum_{i=1}^{2} (x_i \eta_i), \tag{8}$$

where x and η are mole fraction and dynamic viscosity of component i, respectively. The subscripts are as defined in Equation (2).

Molar refraction deviations (ΔR_{m}) were calculated through the following relation:

$$\Delta R_{\mathrm{m}} = \frac{\sum_i x_i M_i}{\rho_{\mathrm{m}}} \frac{n_{\mathrm{D}}^2 - 1}{n_{\mathrm{D}}^2 + 2} - \sum_i x_i \frac{M_i}{\rho_i} \frac{n_{\mathrm{D}i}^2 - 1}{n_{\mathrm{D}i}^2 + 2}, \tag{9}$$

where x, M, ρ, n_{D} are mole fraction, molar mass, density and refractive index, respectively. The subscripts i and m represent pure components and mixture, respectively.

Table 3. Coefficients of Equation (5) of the pure components at $T = 298.15$–$313.15\,\mathrm{K}$.

	A_1	A_2	A_3
THF	909.312	−0.068	−0.001
1-Hexanol	832.437	−0.634	−0.001
1-Heptanol	834.285	−0.572	−0.002
1-Octanol	829.864	−0.045	−0.011
1-Nonanol	840.208	−0.594	−0.001
1-Decanol	848.499	−0.969	0.004

Table 4. Isobaric expansion coefficients of pure components at $T = 298.15$–$313.15\,\mathrm{K}$.

	α_i^{o} ($k\,\mathrm{K}^{-1}$)			
	$T = 298.15\,\mathrm{K}$	$T = 303.15\,\mathrm{K}$	$T = 308.15\,\mathrm{K}$	$T = 313.15\,\mathrm{K}$
THF	1.241	1.255	1.269	1.283
1-Hexanol	0.862	0.883	0.904	0.925
1-Heptanol	0.823	0.852	0.881	0.910
1-Octanol	0.826	0.845	0.865	0.884
1-Nonanol	0.810	0.839	0.853	0.875
1-Decanol	0.821	0.830	0.838	0.847

Values of V^E, $\Delta\eta$, k_s^E, u^D and ΔR_m have been fitted by the method of least squares to a Redlich–Kister polynomial-type equation [10],

$$Y = x_1x_2\sum_{j=1}^{n} a_{j-1}(x_1 - x_2)^{j-1}, \tag{10}$$

where Y refers to the property, and x_1 and x_2 are the mole fractions of pure components 1 and 2, respectively. a_{j-1} are the polynomial coefficients, and n is their number. The correlated results in each case are shown in Table 5, in which the tabulated SD σ, was defined as:

$$\sigma = \left\{\frac{\left[\sum (Y_{\mathrm{exp}} - Y_{\mathrm{cal}})^2\right]}{(N-n)}\right\}^{1/2}, \tag{11}$$

where N is the number of data points and n is the number of the coefficients. Y_{exp} and Y_{cal}, denote the experimental and calculated values of Y, respectively.

The excess molar volume (V^E) for the binary mixtures at 298.15 K is shown in Figure 1. All the five systems exhibit a positive deviation, with an increasing in the deviation with the molecular size of 1-alkanols from hexanol to decanol. This may indicate that when the mixtures are created the excess free volume increases with increasing chain length of 1-alkanols owing to the decrease in their polarisability [26].

Figure 2 shows the equimolar excess molar volumes $V^E(x_1 = 0.5)$, of THF + 1-alkanols mixtures, against number of carbon atoms of 1-alkanols (n). The results show that the deviation of V^E increases with the increase of chain length of 1-alkanols, and so it does with the increase in temperature.

The comparison of V^E ($x_1 = 0.5$) values for the binary mixtures of aromatic ether, (anisole +1-hexanol, or 1-heptanol, or 1-octanol) studied by Weng [27] with corresponding values for the binary mixtures of the cyclic ether (THF + 1-hexanol, or 1-heptanol or 1-octanol) in the present work at 303.15 and 313 K, shows a similar trend of behaviour for both studies.

The values of $\Delta\eta$, ΔR_m, u^D, k_s^E, and their representations by Equation (10) are plotted against x_1 in Figures 3–6.

The deviations of the speed of sound (u^D) and molar refraction deviation (ΔR_m) at 298.15 K over the whole composition range for all the mixtures, are positive and show a systematic trend with the size of the 1-alkanols in the sequence: 1-decanol > 1-nonanol > 1-octanol > 1-heptanol > 1-hexanol. While the deviations in viscosity $\Delta\eta$ and the excess isentropic compressibilities (k_s^E), respectively displayed in Figures 3 and 6, are negative and exhibit a systematic trend with the size of the 1-alkanols. The k_s^E and ΔR_m data of the five binary mixtures shows a maximum in the following order: 1-decanol > 1-nonanol > 1-octanol > 1-heptanol > 1-hexanol.

With increasing temperature the values of k_s^E increases, while u^D values decrease with the same dependencies and systematic variation. The curves are not presented to avoid overcrowding.

The speeds of sound have been estimated for the present binary mixtures at 298.15 K, using the CFT and the FLT. The experimental data of speed of sound at equimolar compositions in all binary mixtures at 298.15 K are compared with the corresponding theoretical values of speed of sound u_{FLT} and u_{CFT} in Table 6. The values of SDs σ,

Table 5. Coefficients of the Redich–Kister equation and SDs σ, of excess and deviation functions for the binary mixtures from $T = 298.15$–313.15 K.

	a_0	a_1	a_2	a_3	σ	a_0	a_1	a_2	a_3	σ
				THF (1) + 1-hexanol (2)						
			$T = 298.15$ K					$T = 303.15$ K		
V^E (cm^3 mol^{-1})	0.1437	−0.0669	0.0087	0.0181	0.0003	0.1388	−0.0777	−0.0059	0.0529	0.0007
$\Delta\eta$ (mPa s)	−5.1955	−2.2902	−0.7708	−0.1335	0.0003	−4.2556	−1.7994	−0.5252	0.0661	0.0004
k_s^E (TPa^{-1})	−32.112	14.735	−0.292	4.308	0.0001	−31.224	15.913	2.820	−2.238	0.0002
u^D (m s^{-1})	28.581	−14.063	−0.207	−2.784	0.0001	27.193	−13.979	−3.417	2.707	0.0007
ΔR_m (cm^3 mol^{-1})	0.1232	0.0215	−0.0077	−0.0191	0.0002	0.1479	0.0007	−0.0130	0.0061	0.0004
			$T = 308.15$ K					$T = 313.15$ K		
V^E (cm^3 mol^{-1})	0.1338	−0.0840	−0.0259	0.0727	0.0008	0.1284	−0.0911	−0.0429	0.1019	0.0008
$\Delta\eta$ (mPa s)	−3.3986	−1.6337	−0.7445	−0.2070	0.0003	−2.8172	−1.3104	−0.4386	0.0178	0.0004
k_s^E (TPa^{-1})	−30.398	17.694	8.134	−7.885	0.0008	−29.204	19.391	9.901	−12.009	0.0010
u^D (m s^{-1})	26.190	−14.527	−7.212	6.962	0.0004	25.106	−12.380	−8.225	9.497	0.0001
ΔR_m (cm^3 mol^{-1})	0.1988	−0.0297	−0.0162	0.0005	0.0007	0.2076	−0.0080	0.0425	−0.0840	0.0008
				THF (1) + 1-heptanol (2)						
			$T = 298.15$ K					$T = 303.15$ K		
V^E (cm^3 mol^{-1})	0.2952	−0.1383	0.0054	0.0059	0.0006	0.2862	−0.1420	−0.0197	0.0279	0.0004
$\Delta\eta$ (mPa s)	−6.9391	−3.3902	−1.8232	−0.9598	0.0004	−5.6809	−2.4644	−0.8632	−0.2462	0.0006
k_s^E (TPa^{-1})	−32.047	21.627	−16.277	22.351	0.0008	−32.858	20.773	−8.180	1.433	0.0007
u^D (m s^{-1})	31.154	−13.751	9.592	−11.383	0.0004	30.490	−14.477	7.370	−3.798	0.0007
ΔR_m (cm^3 mol^{-1})	0.2131	−0.0853	−0.0823	0.0984	0.0006	0.2236	−0.0661	0.0154	0.0282	0.0008
			$T = 308.15$ K					$T = 313.15$ K		
V^E (cm^3 mol^{-1})	0.2764	−0.1566	− 0.0363	0.0831	0.0007	0.2673	−0.1649	−0.0644	0.1440	0.0003
$\Delta\eta$ (mPa s)	−4.4556	−2.9665	−1.1179	0.3221	0.0006	−3.7016	−1.5513	−0.8448	−0.5328	0.0003
k_s^E (TPa^{-1})	−31.508	22.822	−4.480	−6.413	0.0006	−29.661	23.799	−0.792	−11.837	0.0004
u^D (m s^{-1})	28.920	−15.502	7.932	2.875	0.0009	27.264	−16.902	3.593	7.620	0.0003
ΔR_m (cm^3 mol^{-1})	0.2505	−0.0617	0.0268	−0.0223	0.0008	0.2727	−0.0652	0.0706	−0.0467	0.0007
				THF (1) + 1-octanol (2)						
			$T = 298.15$ K					$T = 303.15$ K		
V^E (cm^3 mol^{-1})	0.4731	−0.2319	−0.0515	0.0750	0.0009	0.4504	−0.2550	−0.1026	0.1788	0.0008
$\Delta\eta$ (mPa s)	−9.1159	−4.3494	−1.1759	0.4329	0.0005	−7.2948	−3.3127	−0.8408	0.1418	0.0009

k_s^E (TPa^{-1})	−35.952	20.113	−16.044	9.298	0.0007	−34.383	22.000	−8.426	4.895	0.0006
u^D (m s^{-1})	33.989	−16.222	20.452	−11.025	0.0006	32.748	−21.486	15.983	6.329	0.0005
ΔR_m (cm^3 mol^{-1})	0.2178	−0.0068	−0.0316	−0.0400	0.0007	0.2429	−0.0179	0.0231	−0.0621	0.0009
			$T = 308.15$ K					$T = 313.15$ K		
V^E (cm^3 mol^{-1})	0.4294	−0.2679	−0.1631	0.2319	0.0008	0.4058	−0.2994	−0.2277	0.3612	0.0007
$\Delta\eta$ (mPa s)	−6.0357	−3.4216	−0.1125	1.8693	0.0004	−4.6504	−2.1511	−0.7229	0.2781	0.0004
k_s^E (TPa^{-1})	−32.241	27.891	−5.177	−19.708	0.0005	−31.244	27.212	3.235	−24.082	0.0004
u^D (m s^{-1})	31.791	−18.497	8.830	3.708	0.0006	29.734	−23.427	4.606	18.807	0.0004
ΔR_m (cm^3 mol^{-1})	0.2663	0.0011	0.0665	−0.1486	0.0007	0.2790	0.0204	0.1323	−0.2386	0.0003
					THF (1) + 1-nonanol (2)					
			$T = 298.15$ K					$T = 303.15$ K		
V^E (cm^3 mol^{-1})	0.5992	−0.2235	0.0258	−0.0211	0.0007	0.5740	−0.2260	−0.0442	0.0306	0.0008
$\Delta\eta$ (mPa s)	−11.627	−5.8228	−3.1049	−0.7148	0.0006	−8.9257	−3.6191	−1.8099	−1.1058	0.0005
k_s^E (TPa^{-1})	−37.728	19.611	−24.314	25.338	0.0009	−36.420	26.190	−20.210	2.334	0.0006
u^D (m s^{-1})	36.324	−15.493	22.641	−32.678	0.0001	34.441	−21.159	17.662	−13.421	0.0008
ΔR_m (cm^3 mol^{-1})	0.2402	−0.0558	0.0775	−0.1020	0.0005	0.2927	−0.0876	0.1285	−0.1800	0.0005
			$T = 308.15$ K					$T = 313.15$ K		
V^E (cm^3 mol^{-1})	0.5493	−0.2676	−0.0977	0.1731	0.0007	0.5158	−0.3079	−0.1543	0.3530	0.0005
$\Delta\eta$ (mPa s)	−7.0792	−3.4078	−1.8692	−0.7289	0.0036	−5.6401	−2.2911	−0.8119	0.0287	0.0005
k_s^E (TPa^{-1})	−36.394	26.282	−20.629	2.519	0.0007	−32.075	34.300	−17.526	−18.620	0.0009
u^D (m s^{-1})	33.412	−26.675	13.285	6.084	0.0009	31.410	−28.700	10.535	22.241	0.0006
ΔR_m (cm^3 mol^{-1})	0.3335	−0.1158	0.1655	−0.1915	0.0003	0.3759	−0.0942	0.2268	−0.3263	0.0004
					THF (1) + 1-decanol (2)					
			$T = 298.15$ K					$T = 303.15$ K		
V^E (cm^3 mol^{-1})	0.8699	−0.2155	0.0052	−0.1318	0.0002	0.8443	−0.2415	−0.0900	0.0909	0.0009
$\Delta\eta$ (mPa s)	−14.207	−5.5701	−1.1419	−0.0072	0.0005	−11.146	−4.5465	−2.2199	−1.1412	0.0005
k_s^E (TPa^{-1})	−38.802	19.497	−32.930	52.320	0.0008	−39.643	28.448	−20.387	10.883	0.0005
u^D (m s^{-1})	41.790	−37.210	23.951	−13.760	0.0004	35.947	−20.181	30.431	−38.492	0.0007
ΔR_m (cm^3 mol^{-1})	0.2541	−0.0279	0.0079	−0.1019	0.0009	0.2844	−0.0281	0.0999	−0.3013	0.0006
			$T = 308.15$ K					$T = 313.15$ K		
V^E (cm^3 mol^{-1})	0.8091	−0.3033	−0.1739	0.3386	0.0007	0.7795	−0.2985	−0.3165	0.4122	0.0004
$\Delta\eta$ (mPa s)	−8.7022	−3.5916	−1.5214	−0.4650	0.0002	−6.9060	−2.6719	−1.1060	−0.3334	0.0007
k_s^E (TPa^{-1})	−37.501	31.399	−18.258	2.645	0.0003	−36.157	34.813	−15.290	−13.664	0.0004
u^D (m s^{-1})	34.714	−32.870	23.626	3.317	0.0009	33.348	−26.207	14.228	−0.4953	0.0006
ΔR_m (cm^3 mol^{-1})	0.3090	−0.0488	0.1522	−0.3109	0.0004	0.3540	0.0667	0.0919	−0.6633	0.0005

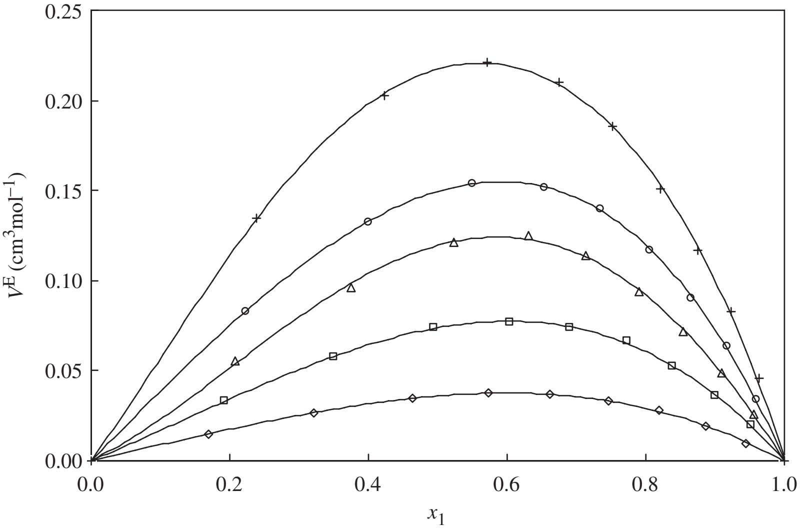

Figure 1. Excess molar volumes (V^E) for the binary mixtures: ◇, THF + hexanol; □, +heptanol; Δ, +octanol; ○, +nonanol; +, +decanol at 298.15 K; solid line (Redlich–Kister).

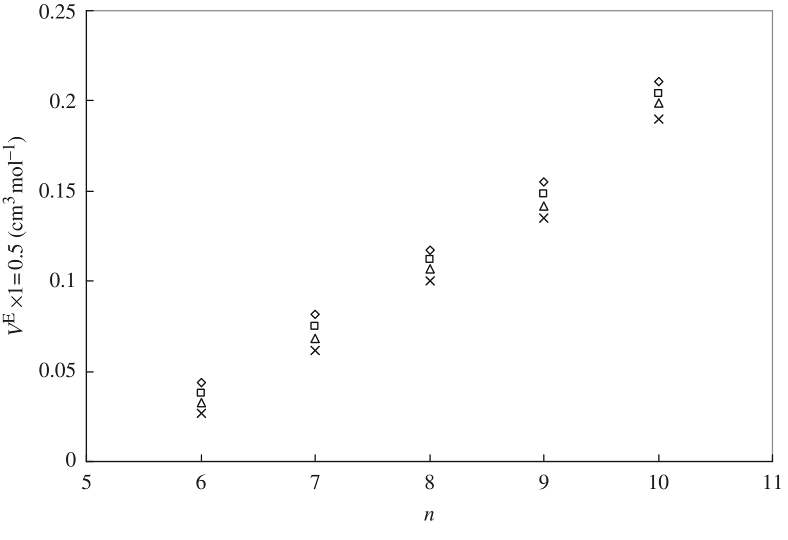

Figure 2. Equimolar excess molar volume (V^E), against the number of carbon atoms n of 1-alkanols for binary mixtures of THF (1) + 1-alkanol (2) at: ◇, 298.15 K; □, 303.15 K; Δ, 308.15 K; x, 313.15 K.

for each individual mixture are also included in Table 6. The analysis of standard percentage deviations of u_{CFT} and u_{FLT} reveals that their values are in the range from 0.06 to 1.04 and from 2.24 to 3.40, with average values of 0.42 and 2.93, respectively. These values show that the CFT of Schaaffs is more appropriate for prediction of sound speed data for presently investigated mixtures.

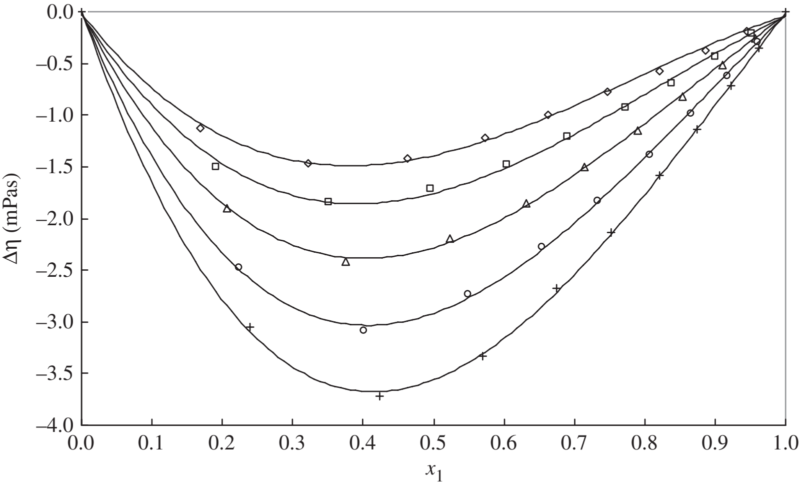

Figure 3. Viscosity deviation $\Delta\eta$, for the binary mixtures at 298.15 K. Symbols are the same as those given in Figure 1.

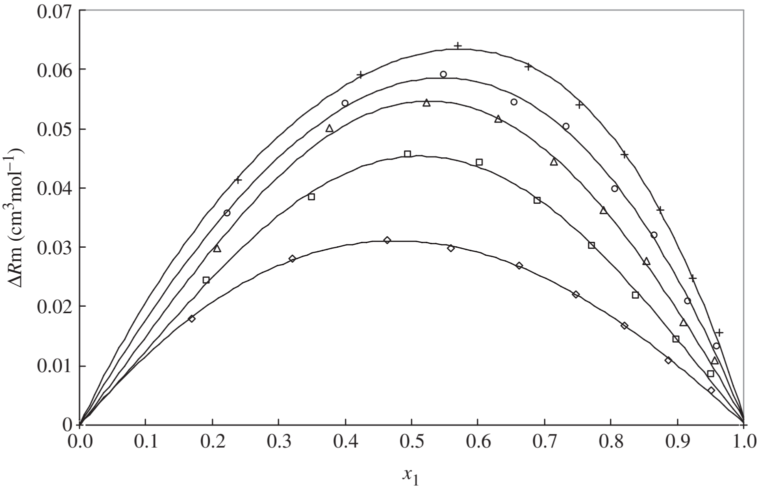

Figure 4. Molar refraction deviation ΔR_m for the binary mixtures at 298.15 K. Symbols are the same as those given in Figure 1. Solid lines correspond to R_m calculated by Lorentz–Lorsnz equation.

The results obtained for viscosity of binary mixtures were used to test the semi-empirical relations of the three-body and the four-body McAllister as reported in our previous work [7]. The correlation parameters and SDs for the proposed equations are shown in Table 7. The average SDs show that these relations predict viscosities adequately for the systems under study. The best correlation method giving, relatively, lowest SD is found to be the McAllister four-body equation, with SD (σ) range from 0.0014 to 0.0079.

The experimental refractive index data (n_{Dexp}) of THF + 1-alkanol binary mixtures at temperature range from 298.15 to 313.15 K were compared with the corresponding calculated values (n_{Dcal}) using the Lorentz–Lorenz and Gladstone–Dale equations with

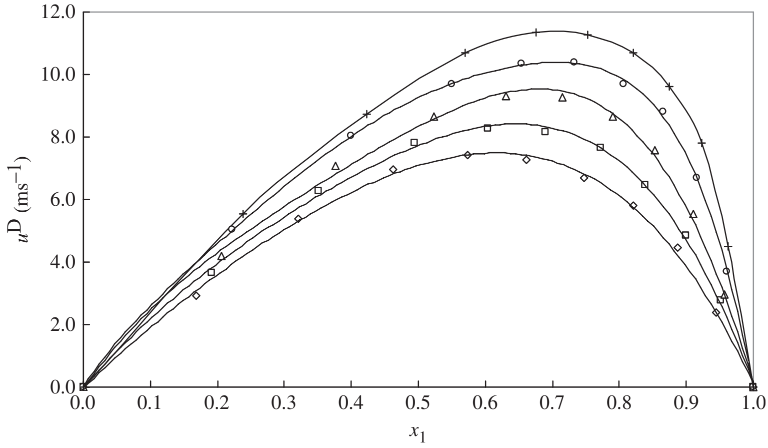

Figure 5. Deviation of the speeds of sound u_D from their ideal values for the binary mixtures: at 298.15 K. Symbols are the same as those given in Figure 1. Solid line (Redlich–Kister).

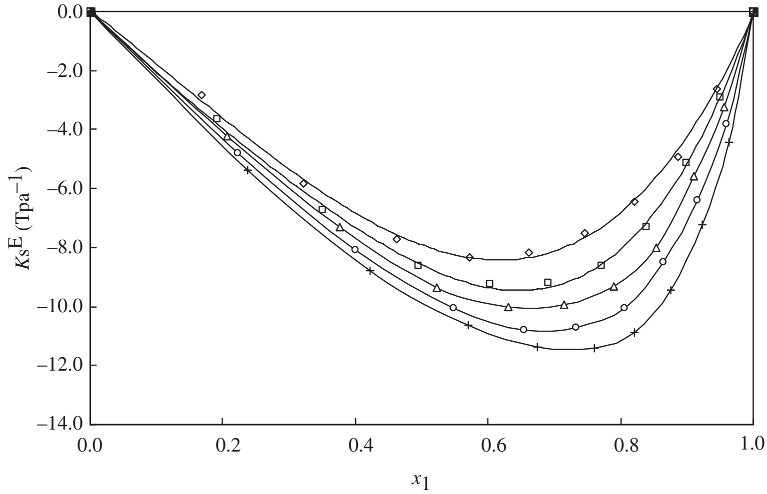

Figure 6. Deviation in isentropic compressibility k_s^E *vs*. mole fraction of THF at 298.15 K. Symbols are the same as those given in Figure 1. Solid line (Redlich–Kister).

Table 6. Comparison of speeds of sound estimated from the CFT and FLT with those observed experimentally for the binary mixtures at 298.15 K.

	u_{exp} (m s^{-1})	u_{CFT} (m s^{-1})	σ (%)	u_{FLT} (m s^{-1})	σ (%)
THF (1) + 1-hexanol	1287.02	1286.60	0.06	1331.76	3.40
THF (1) + 1-heptanol	1300.53	1298.27	0.07	1335.43	2.82
THF (1) + 1-octanol	1312.05	1316.72	0.09	1354.03	2.24
THF (1) + 1-nonanol	1330.22	1333.59	1.04	1350.83	3.15
THF (1) + 1-decanol	1346.85	1342.50	0.86	1361.80	3.05
Average standard percentage deviation			0.42		2.93

Table 7. Values of parameters and SD of viscosity equations for binary mixtures at different temperatures.

	ν_{12}	ν_{21}	ν_{1122}	ν_{1112}	ν_{2221}	σ
THF (1) + 1-hexanol (2)						
$T = 298.15\,K$						
McAllister three-body	0.864	2.245				0.0051
McAllister four-body			0.748	1.412	2.033	0.0045
$T = 303.15\,K$						
McAllister three-body	0.770	2.025				0.0070
McAllister four-body			0.677	1.295	1.772	0.0072
$T = 308.15\,K$						
McAllister three-body	0.843	1.682				0.0041
McAllister four-body			0.703	1.218	1.272	0.0038
$T = 313.15\,K$						
McAllister three-body	0.789	1.542				0.0065
McAllister four-body			0.670	1.100	1.382	0.0051
THF (1) + 1-heptanol (2)						
$T = 298.15\,K$						
McAllister three-body	1.081	2.630				0.0040
McAllister four-body			0.884	1.742	3.304	0.0031
$T = 303.15\,K$						
McAllister three-body	0.961	2.513				0.0032
McAllister four-body			0.807	1.578	3.108	0.0030
$T = 308.15\,K$						
McAllister three-body	1.356	1.693				0.0060
McAllister four-body			1.018	1.508	2.255	0.0051
$T = 313.15\,K$						
McAllister three-body	0.925	1.947				0.0033
McAllister four-body			0.743	1.376	2.366	0.0026
THF (1) + 1-octanol (2)						
$T = 298.15\,K$						
McAllister three-body	1.085	2.969				0.0088
McAllister four-body			0.854	1.869	3.639	0.0079
$T = 303.15\,K$						

(*continued*)

Table 7. Continued.

	ν_{12}	ν_{21}	ν_{1122}	ν_{1112}	ν_{2221}	σ
McAllister three-body	1.223	3.059				0.0030
McAllister four-body			0.986	1.866	3.942	0.0019
$T = 308.15$ K						
McAllister three-body	1.231	2.582				0.0058
McAllister four-body			0.969	1.675	3.395	0.0041
$T = 313.15$ K						
McAllister three-body	1.152	2.412				0.0015
McAllister four-body			0.882	1.674	2.966	0.0014
THF (1) + 1-nonanol (2)						
$T = 298.15$ K						
McAllister three-body	1.642	4.262				0.0035
McAllister four-body			1.194	2.785	5.260	0.0036
$T = 303.15$ K						
McAllister three-body	1.475	3.923				0.0047
McAllister four-body			1.090	2.527	4.711	0.0030
$T = 308.15$ K						
McAllister three-body	1.647	3.033				0.0021
McAllister four-body			1.174	2.212	3.882	0.0016
$T = 313.15$ K						
McAllister three-body	1.423	2.975				0.0079
McAllister four-body			1.023	2.104	3.591	0.0070
THF (1) + 1-decanol (2)						
$T = 298.15$ K						
McAllister three-body	1.879	5.827				0.0030
McAllister four-body			1.352	3.280	7.274	0.0022
$T = 303.15$ K						
McAllister three-body	1.804	4.777				0.0067
McAllister four-body			1.256	3.135	5.662	0.0044
$T = 308.15$ K						
McAllister three-body	1.600	3.258				0.0049
McAllister four-body			1.112	2.229	4.128	0.0047
$T = 313.15$ K						
McAllister three-body	1.781	3.498				0.0045
McAllister four-body			1.232	2.431	4.395	0.0040

average deviations of 18×10^{-5}, 26×10^{-5}, 19×10^{-5}, 24×10^{-5} and 28×10^{-5}, 33×10^{-5}, 29×10^{-5}, 37×10^{-5} at 298.15, 303.15, 308.15 and 313.15 K, respectively. The mixing rule proposed by Lorentz–Lorenz equation is more suitable for prediction of our data.

4. Conclusions

In this work, we have measured the liquid densities, viscosities, refractive indices and speeds of sound of THF + 1-hexanol, or 1-heptanol, or 1-octanol, or 1-nonanol, or 1-decanol binary mixtures at temperatures between 298.15 and 313.15 K. Excess molar volume, deviation in speed of sound, excess molar isentropic compressibility, deviations in the molar refractivity and viscosity deviation for binary mixtures have been calculated and fitted to a Redlich–Kister equation. The excess molar volume, deviation in speed of sound and molar refraction deviation showed a positive behaviour, while the excess isentropic compressibility and the deviation in viscosity exhibited negative behaviour for the systems under investigation. Examination of the standard percentage deviation between the experimental and the theoretical values of speed of sound showed a better prediction using the CFT. The four-body McAllister equation provided more adequate representation of viscosities data than other semi-empirical relation tested for the present binary mixtures.

Acknowledgement

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