



Studies on liquid–liquid interactions of some ternary mixtures by density, viscosity, ultrasonic speed and refractive index measurements

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ABSTRACT

The excess molar volumes, V^E , and viscosity deviations, $\Delta\eta$, were calculated from the measured density and viscosity data over the whole composition range for the ternary systems of 1,3-dioxolane (DO), dichloromethane (DM) + methyl acetate (MA), ethyl acetate (EA), propyl acetate (PA), butyl acetate (BA) and isoamyl acetate (IA) at 298.15 K. From the experimental observations the speed of sounds and refractive indices of these ternary mixtures have been measured over the entire range of composition at the same temperature. Also the isentropic compressibility, deviation in isentropic compressibility and deviation in molar refraction have been evaluated. The excess or deviation properties were found to be either negative or positive depending on the molecular interactions and the nature of liquid mixtures.

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1. Introduction

Densities and viscosities of liquids and liquid mixtures are essential for many engineering and industrial applications. The mixture functions such as the excess molar volume (V^E), the viscosity deviation ($\Delta\eta$), deviation in isentropic compressibility (ΔK_S), and deviation in molar refraction (ΔR), are often used to describe the intermolecular forces in mixtures to help us understand their real behavior and to develop models for their description as well as simulation processes. Therefore, over the years, studies on multicomponent liquid systems have attracted the attention of many researchers [1–6].

To the best of our knowledge, the properties of mixtures of these liquids have not been reported earlier. In the present paper, we report density (ρ), viscosity (η), speed of sound (u) and refractive index (n_D) for the ternary systems consisting of 1,3-dioxolane (DO), dichloromethane (DM) + methyl acetate (MA), ethyl acetate (EA), propyl acetate (PA), butyl acetate (BA) and isoamyl acetate (IA) at 298.15 K and atmospheric pressure over the entire composition range. The experimental data are used to calculate excess molar volumes (V^E), deviations in viscosity ($\Delta\eta$), deviations in isentropic compressibility (ΔK_S) and deviation in the molar refraction ΔR of the mixtures at 298.15 K under atmospheric pressure.

2. Experimental

2.1. Chemicals

High-purity spectroscopic and analytical grade samples of DO, MA, EA, PA, BA and IA were procured from S.D. Fine Chemicals Ltd., Mumbai, India. DM was purchased from Sisco Research Laboratories Pvt. Ltd., Mumbai, India. All the samples were used without further purification because their purities exceeded 99% as tested by gas chromatography (HP 6890 series) using a flame ionization detector with a packed column. DM (A.R.) was fractionally distilled over calcium oxide, and the middle colorless fractions were collected. Experimental values of η and n_D of the pure liquids are compared in Table 1 [7–12] at 298.15 K and these values agree well with the published results. Mixtures were prepared by mass in specially designed glass stoppered bottles and were used on the same day. The weights were taken on a Mettler electronic analytical balance (AG285).

2.2. Measurements

Densities were measured with an Ostwald-Sprengel-type pycnometer having a bulb volume of about 25 cm³ and an internal diameter of the capillary of about 0.1 cm. The pycnometer was calibrated at 298.15 K with doubly distilled water. The pycnometer with experimental liquid was equilibrated in a glass-walled thermostated water bath maintained at ± 0.01 K of the desired temperature. The pycnometer was then removed from the thermostat, properly dried, and weighed in an electronic balance with

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Table 1
Physical properties of pure solvents at 298.15 K.

Components	$\rho \times 10^{-3}$ (kg m ⁻³)		η (mPa s)		u (m s ⁻¹)		n_D	
	Exp	lit	Exp	Lit	Exp	Lit	Exp	Lit
1,3-Dioxolane	1.0570	1.0577 [7]	0.588	0.5878 [7]	1339	1338.8 [7]	1.398	1.3983 [11]
Dichloromethane	1.3180	1.3163 [8]	0.380	0.380 [8]	1035	1035 [8]	1.422	1.4228 [8]
Methyl acetate	0.9282	0.92680 [10]	0.374	0.384 [10]	1155	1150 [10]	1.446	1.3606 [9]
Ethyl acetate	0.8928	0.89455 [10]	0.428	0.428 [10]	1142	1138 [10]	1.371	1.3710 [11]
Propyl acetate	0.8823	0.8831 [11]	0.550	0.551 [11]	1179	1172 [11]	1.384	1.3835 [11]
Butyl acetate	0.8756	0.87619 [10]	0.660	0.674 [10]	1201	1190 [10]	1.393	1.3931 [11]
Isoamyl acetate	0.8671	0.86621 [10]	0.780	0.781 [10]	1194	1195 [10]	1.398	1.39836 [12]

a precision of ± 0.01 mg. Adequate precautions were taken to avoid evaporation losses during the time of measurements. An average of triplicate measurement was taken into account. The estimated uncertainty for V^E is within 0.001–0.014 cm³ mol⁻¹. The details of the methods and measurement techniques had been described earlier [13].

The viscosity was measured by means of a suspended Ubbelohde type viscometer, calibrated at 298.15 K with triply distilled water and purified methanol using density and viscosity values from the literature [14,15]. The flow times were accurate to ± 0.1 s, and the uncertainty in the viscosity measurements, based on our work on several pure liquids, is ± 0.003 mPa s. The details of the methods and techniques have been described earlier [13].

Speeds of sound were determined by a multi-frequency ultrasonic interferometer (Mittal Enterprise, New Delhi) working at 1 MHz, which was calibrated with water, methanol and benzene at 298.15 K. The precision of the speed measurements was ± 0.2 m s⁻¹. The details of the methods and techniques have been described earlier [16].

The refractive indices of pure liquids and their binary mixture were measured by using a thermostated Abbe refractometer. The refractometer was calibrated by measuring the refractive indices of triply distilled water and toluene at desired temperatures. The values of refractive indices were obtained using sodium D light. The uncertainty of refractive index measurements was within 0.0002. All measurements described above were performed at least three times at atmospheric pressure and an average of at least three measurements was calculated for each temperature.

3. Results

The experimentally determined density values along with the excess molar volumes, V^E are listed in Table 2. The excess molar volumes, V^E , are calculated from density data according to the following equation [17]:

$$V^E = \sum_{i=1}^3 x_i M_i \left(\frac{1}{\rho} - \frac{1}{\rho_i} \right) \quad (1)$$

where ρ is the density of the mixture and M_i , x_i , and ρ_i are the molecular weight, mole fraction, and density of the i th component in the mixture, respectively.

In Table 2, the experimentally determined viscosities, speeds of sound, isentropic compressibilities, refractive indices and molar refractivities of the ternary mixtures are presented along with the mole fraction of DO, x_A , and DM, x_B , at the experimental temperature. Quantitatively, as per the absolute reaction rates theory [18], the deviations of viscosities can be calculated as

$$\Delta\eta = \eta - \sum_{i=1}^3 (x_i \eta_i) \quad (3)$$

where η is the dynamic viscosity of the mixture and x_i and η_i are the mole fraction and viscosity of the i th component in the mixture, respectively.

Isentropic compressibilities (K_S), and the deviations in isentropic compressibilities (ΔK_S), are calculated from the experimental densities (ρ), and speeds of sound (u), using the following equations [19]

$$K_S = \frac{1}{(u^2 \rho)} \quad (5)$$

$$\Delta K_S = K_S - \sum_{i=1}^3 (x_i K_{S,i}) \quad (6)$$

where u is the speed of sound of the mixture and x_i and $K_{S,i}$ are the mole fraction and isentropic compressibility of the i th component in the mixture, respectively.

The molar refractivity (R) is calculated using the Lorentz–Lorenz equation [20]

$$R = \left[\frac{(n_D^2 - 1)}{(n_D^2 + 2)} \right] V \quad (7)$$

where n_D and V being the refractive indices and the molar volume, respectively.

The deviation in the molar refraction ΔR was calculated on a mole fraction basis as suggested by Brocos et al. [21]

$$\Delta R = R - \sum_{i=1}^3 (x_i R_i) \quad (8)$$

where R is the molar refraction of the mixture and x_i and R_i are the mole fraction and molar refraction of the i th component in the mixture, respectively.

4. Discussion

A perusal of Table 2 shows that for the studied ternary systems of DO + DM + alkyl acetates, the V^E values are shifted from positive to negative as the alkyl chain length of the alkyl acetates increases. The negative V^E values may be attributed to specific interactions [22,23] among the mixing components in the mixtures, the positive values often suggest dominance of dispersion forces [22,23] among them. Therefore, specific interactions for the alkyl acetates in the studied ternary systems with DO and DM follow the order

$$IA > BA > PA > EA > MA$$

Fig. 1 represents the excess molar volume, V^E , for the five ternary mixtures under taken as a function of mole fraction of DO, x_A . The investigated ternary mixtures show the minima at the mole fraction range, $x_A = 0.6$ –0.7. The V^E values shift from positive to negative as the amount of DO increases in the mixture, which indicates unlike molecular interactions in this system for all the ternary mixtures.

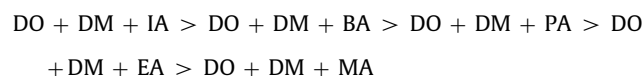
Table 2
Experimental densities (ρ) and excess molar volumes (V^E), experimental viscosities (η_{exp}), speeds of sound (u), isentropic compressibilities (K_S), refractive indices (n_D) and molar refractivities (R) of ternary mixtures of 1,3-dioxolane + dichloromethane + alkyl acetates at 298.15 K.

x_A	x_B	$\rho_{\text{exp}} \times 10^{-3}$ (kg m ⁻³)	$V^E \times 10^6$ (m ³ mol ⁻¹)	η_{exp} (mPa s)	u (m s ⁻¹)	$K_S \times 10^{12}$ (Pa ⁻¹)	n_D	$R \times 10^6$ (m ³ mol ⁻¹)
1,3-Dioxolane + dichloromethane + methyl acetate								
0.0000	0.4659	1.0780	0.760	0.425	1056	831	1.380	16.996
0.1061	0.4165	1.0777	0.558	0.439	1089	783	1.382	16.969
0.2108	0.3677	1.0770	0.379	0.453	1123	737	1.384	16.944
0.3140	0.3196	1.0759	0.233	0.469	1155	697	1.386	16.918
0.4159	0.2721	1.0744	0.116	0.484	1186	662	1.387	16.895
0.5165	0.2253	1.0725	0.028	0.500	1215	632	1.389	16.876
0.6157	0.1790	1.0701	-0.031	0.518	1239	608	1.391	16.863
0.7137	0.1334	1.0671	-0.043	0.535	1263	587	1.392	16.857
0.8104	0.0884	1.0638	-0.031	0.552	1287	568	1.394	16.862
0.9058	0.0439	1.0603	-0.011	0.571	1311	549	1.396	16.880
1.0000	0.0000	1.0570	0.000	0.588	1339	528	1.398	16.915
1,3-Dioxolane + dichloromethane + ethyl acetate								
0.0000	0.5092	1.0592	0.411	0.456	1059	843	1.388	19.287
0.1148	0.4507	1.0596	0.312	0.467	1088	797	1.389	19.004
0.2259	0.3941	1.0602	0.208	0.480	1126	745	1.390	18.729
0.3335	0.3394	1.0607	0.115	0.493	1162	698	1.391	18.463
0.4377	0.2863	1.0610	0.038	0.507	1196	659	1.392	18.205
0.5386	0.2349	1.0611	-0.027	0.520	1225	628	1.393	17.958
0.6365	0.1851	1.0609	-0.062	0.534	1250	603	1.394	17.721
0.7315	0.1367	1.0602	-0.066	0.547	1273	582	1.395	17.497
0.8236	0.0898	1.0592	-0.050	0.561	1295	563	1.396	17.290
0.9131	0.0442	1.0580	-0.019	0.575	1316	546	1.397	17.095
1.0000	0.0000	1.0570	0.000	0.588	1339	528	1.398	16.915
1,3-Dioxolane + dichloromethane + propyl acetate								
0.0000	0.5460	1.0535	0.289	0.518	1181	813	1.397	21.193
0.1221	0.4793	1.0542	0.231	0.522	1114	764	1.397	20.665
0.2384	0.4158	1.0552	0.143	0.528	1148	719	1.397	20.154
0.3492	0.3553	1.0562	0.060	0.536	1187	672	1.397	19.667
0.4549	0.2976	1.0572	-0.011	0.543	1218	637	1.397	19.205
0.5559	0.2425	1.0578	-0.060	0.551	1246	609	1.397	18.768
0.6525	0.1897	1.0582	-0.089	0.558	1268	588	1.398	18.354
0.7450	0.1392	1.0584	-0.096	0.567	1288	570	1.398	17.963
0.8335	0.0909	1.0579	-0.065	0.574	1306	554	1.398	17.601
0.9185	0.0445	1.0575	-0.035	0.582	1320	542	1.398	17.254
1.0000	0.0000	1.0570	0.000	0.588	1339	528	1.398	16.915
1,3-Dioxolane + dichloromethane + butyl acetate								
0.0000	0.5777	1.0492	0.268	0.575	1181	788	1.403	22.827
0.1283	0.5035	1.0505	0.187	0.576	1114	745	1.403	22.062
0.2488	0.4340	1.0519	0.100	0.578	1148	699	1.402	21.342
0.3621	0.3685	1.0534	0.020	0.580	1187	656	1.402	20.660
0.4689	0.3068	1.0547	-0.046	0.582	1218	622	1.401	20.019
0.5698	0.2485	1.0558	-0.092	0.583	1246	595	1.401	19.418
0.6652	0.1934	1.0566	-0.111	0.584	1268	574	1.400	18.856
0.7555	0.1412	1.0567	-0.084	0.586	1288	558	1.400	18.335
0.8412	0.0917	1.0568	-0.053	0.586	1306	546	1.399	17.838
0.9226	0.0447	1.0569	-0.028	0.587	1320	536	1.399	17.364
1.0000	0.0000	1.0570	0.000	0.588	1339	528	1.398	16.915
1,3-Dioxolane + dichloromethane + isoamyl acetate								
0.0000	0.6052	1.0433	0.254	0.633	1115	771	1.406	24.213
0.1336	0.5243	1.0454	0.155	0.629	1148	726	1.406	23.232
0.2576	0.4493	1.0474	0.070	0.624	1182	683	1.405	22.320
0.3729	0.3795	1.0495	-0.019	0.620	1217	644	1.404	21.468
0.4806	0.3144	1.0515	-0.091	0.616	1247	612	1.404	20.677
0.5812	0.2535	1.0534	-0.150	0.613	1273	586	1.403	19.939
0.6755	0.1964	1.0547	-0.157	0.609	1296	565	1.402	19.251
0.7640	0.1428	1.0554	-0.126	0.605	1314	549	1.401	18.609
0.8473	0.0924	1.0560	-0.088	0.600	1327	538	1.400	18.008
0.9259	0.0449	1.0565	-0.042	0.593	1333	533	1.399	17.448
1.0000	0.0000	1.0570	0.000	0.588	1339	528	1.398	16.915

This may be due to the differences in molecular sizes of the components of the mixtures, dipole–dipole type forces [24] resulting from the polarisability of ester molecules by the dipoles of the liquid components [25] of the mixtures and donor–acceptor interaction.

Table 2 also shows that the values of experimental viscosities, η_{exp} , increase with increasing of carbon chain length of the alkyl acetates in the ternary mixtures and are depicted in Fig. 2a as a

function of the mole fraction of DO, x_A . The η_{exp} , values are varied almost linearly with the mole fraction of DO, x_A and also the experimental viscosities are higher than the theoretical viscosities. The order it follows is



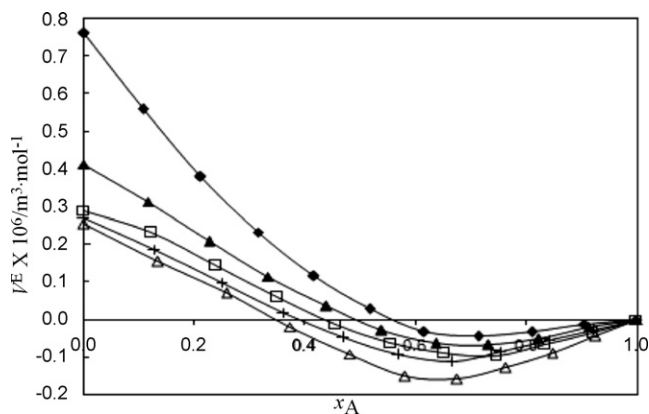


Fig. 1. Excess molar volumes, V^E , of: (◆) 1,3-dioxolane + dichloromethane + methyl acetate; (▲) 1,3-dioxolane + dichloromethane + ethyl acetate; (□) 1,3-dioxolane + dichloromethane + propyl acetate; (+) 1,3-dioxolane + dichloromethane + butyl acetate; (△) 1,3-dioxolane + dichloromethane + isoamyl acetate mixtures with mole fraction of 1,3-dioxolane, x_A , at 298.15 K.

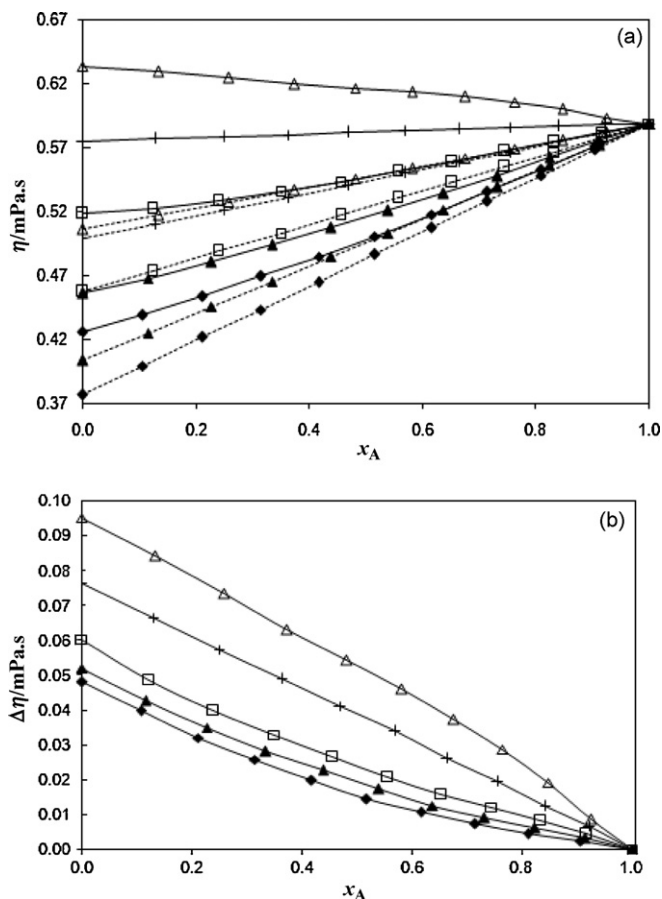


Fig. 2. (a) Theoretical (---) and experimental (—) viscosities, η , of: (◆) 1,3-dioxolane + dichloromethane + methyl acetate; (▲) 1,3-dioxolane + dichloromethane + ethyl acetate; (□) 1,3-dioxolane + dichloromethane + propyl acetate; (+) 1,3-dioxolane + dichloromethane + butyl acetate; (△) 1,3-dioxolane + dichloromethane + isoamyl acetate mixtures with mole fraction of 1,3-dioxolane, x_A , at 298.15 K. (b) Viscosity deviation, $\Delta\eta$, of: (◆) 1,3-dioxolane + dichloromethane + methyl acetate; (▲) 1,3-dioxolane + dichloromethane + ethyl acetate; (□) 1,3-dioxolane + dichloromethane + propyl acetate; (+) 1,3-dioxolane + dichloromethane + butyl acetate; (△) 1,3-dioxolane + dichloromethane + isoamyl acetate mixtures with mole fraction of 1,3-dioxolane, x_A , at 298.15 K.

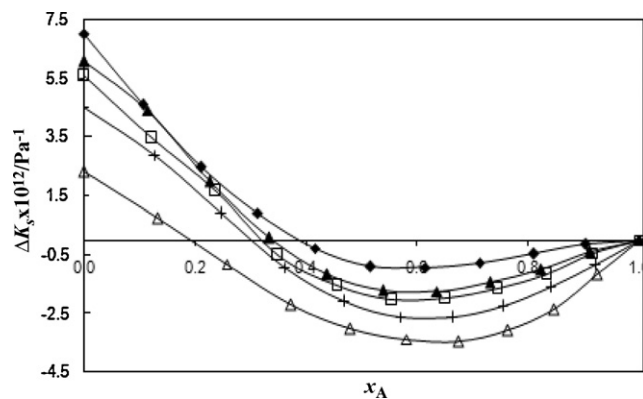
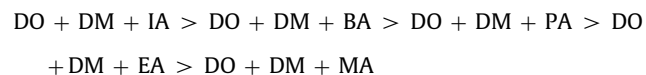


Fig. 3. Deviation in isentropic compressibility, ΔK_S , of: (◆) 1,3-dioxolane + dichloromethane + methyl acetate; (▲) 1,3-dioxolane + dichloromethane + ethyl acetate; (□) 1,3-dioxolane + dichloromethane + propyl acetate; (+) 1,3-dioxolane + dichloromethane + butyl acetate; (△) 1,3-dioxolane + dichloromethane + isoamyl acetate mixtures with mole fraction of 1,3-dioxolane, x_A , at 298.15 K.

As far as $\Delta\eta$ values are concerned, it is positive for all the studied ternary systems over the entire composition range at the experimental temperature. A perusal of Fig. 2b shows that the $\Delta\eta$ values become more positive as the chain length of the alkyl acetates increases. The negative values imply the presence of dispersion forces [26] among the mixing components in the mixtures, and the positive values may be attributed to the presence of specific interactions [26] among them. The $\Delta\eta$ values have been compared for the alkyl acetates with increasing C-chain length which follow the trend



This may also be ascribed due to the complex formation between free electrons of the oxygen and chlorine, different polarisabilities of alkyl acetates, different molar volumes and dipole–dipole interaction. Thus it supports the earlier results obtained from the V^E values.

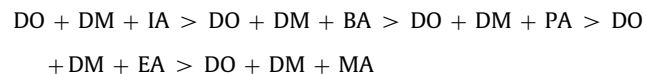
The ΔK_S values are shifted from positive to negative for all the ternary mixtures over the entire range of compositions at 298.15 K, and the deviations in isentropic compressibility, ΔK_S , have been plotted against mole fraction of DO, x_A , for the ternary mixtures at 298.15 K in Fig. 3.

The observed values of ΔK_S can be qualitatively explained by considering the factors, namely (i) the mutual disruption of associates present in the pure liquids, (ii) the formation of weak bonds by due to dipole–dipole and dipole induced dipole [27–29] interaction between the molecules resulting in the formation of electron-transfer complexes and (iii) geometrical fitting of component molecules into each other structure. The first factor contributes to positive ΔK_S values, whereas the remaining two factors lead to negative ΔK_S values.

It is seen that the strength of interaction for the alkyl acetates in studied ternary systems with DO and DM follows the order:



The dependence of molar refraction, ΔR on mole fraction, x_A of DO at 298.15 K is displayed in Fig. 4. It is observed that, for all the mixture ΔR is negative over the whole of the mixture composition and the trend is just reverse of V^E , i.e., the positive ΔR values follow the order



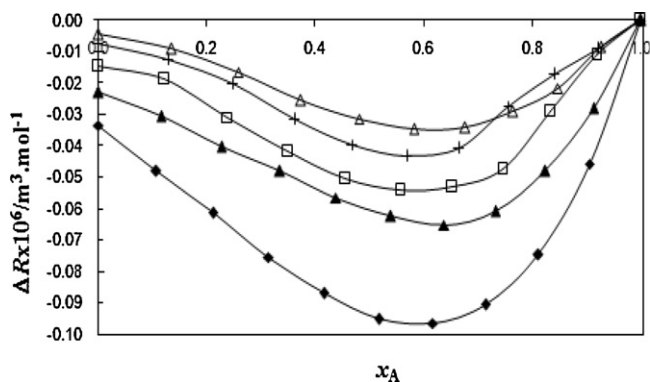


Fig. 4. Deviation in molar refraction (ΔR) of: (◆), 1,3-dioxolane + dichloromethane + methyl acetate; (▲), 1,3-dioxolane + dichloromethane + ethyl acetate; (□), 1,3-dioxolane + dichloromethane + propyl acetate; (+), 1,3-dioxolane + dichloromethane + butyl acetate and (Δ), 1,3-dioxolane + dichloromethane + isoamyl acetate mixtures with mole fraction of 1,3-dioxolane, x_A , at 298.15 K.

The negative ΔR values decrease systematically with decreasing size of the molecules, i.e., from IA to MA. Such values may be due to the electronic perturbation of the individual molecules during mixing and therefore, depend very much on the nature of the mixing components.

5. Conclusion

The investigated mixtures were chosen in order to obtain information about the molecular interactions between their components. After a thorough study of the behavior of 1,3-dioxolane + dichloromethane + alkyl acetates mixtures, we get a clear idea about the type and extent of molecular interactions occurring between the mixture components. The strength of interaction of alkyl acetates with 1,3-dioxolane + dichloromethane mixtures increase with the increasing chain length of the alkyl acetates. The results found in the investigated systems are in accordance with the usual discrepancy in size, polarity of the solvents and donor–acceptor interaction of the components.

List of symbols

x_A	Mole fraction of 1,3-dioxolane
x_B	Mole fraction of dichloromethane
ρ	Density
V^E	Excess molar volume
η	Viscosity
η_{exp}	Experimental viscosity of mixture
$\Delta\eta$	Deviation in viscosity

u	Speed of sound
K_S	Isentropic compressibility
ΔK_S	Deviation in isentropic compressibility
n_D	Refractive index
R	Molar refraction
ΔR	Deviation in molar refraction

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