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Thermodynamic and acoustic properties of binary mixtures of ethers. IV. Diisopropyl ether or oxolane with N,N-dimethylaniline or N,N-diethylaniline at 303.15, 313.15 and 323.15 K

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ABSTRACT

Densities ρ and speeds of sound u, of binary mixtures formed by N,N-dimethylaniline (DMA) or N,N-diethylaniline (DEA) with diisopropyl ether (DIPE) or oxolane have been measured over the entire range of composition at a temperature of (303.15, 313.15 and 323.15 K) and atmospheric pressure. The ρ and u values were used to calculate isentropic compressibilities κ_S, Rao's molar sound functions R, intermolecular free lengths L_f, specific acoustic impedances Z, excess molar volumes V_m^E, excess isentropic compressibilities κ_S^E, excess intermolecular free lengths L_f^E and excess specific acoustic impedances Z^E. The results have been used to investigate molecular interactions and structural effects in these mixtures. The speeds of sound in present mixtures have been estimated using several empirical and theoretical models to determine their relative predicting ability in terms of pure component properties.

1. Introduction

The thermodynamic, acoustic and transport properties of non-electrolyte liquid–liquid mixtures provide information about type and extent of molecular interactions, and can be used for the development of molecular models for describing the behaviour of solutions [1–5].

We are engaged in systematic investigations of binary mixtures of aromatic amine with ether at different temperatures. In the previous papers [6–8] we have reported speeds of sound, isentropic compressibilities, excess molar volumes and various other related properties of binary mixtures of diisopropyl ether (DIPE) or oxolane with aniline, N-methylaniline, N-ethylaniline, o-toluidine and m-toluidine (primary and secondary arylamines) at 303.15, 313.15 and 323.15 K. It was observed that there exist specific interactions through hydrogen bonding and dipole–dipole interactions between primary/secondary arylamine and ether molecules. It would be advantageous to investigate and interpret the volumetric and acoustic results when a tertiary amine is mixed with ether molecules, because in contradiction to primary and secondary arylamines, the tertiary ones cannot form hydrogen bonds with ethers [9–13]. Therefore, in this paper, we extend our studies to the binary mixtures formed from DIPE or oxolane with N,N-dimethylaniline (DMA) or N,N-diethylaniline (DEA). Speeds of sound at different temperatures in the investigated binary mixtures have also been estimated through Nomoto model (NM) [14], Van Dael model (VM) [15], Ernst et al. model (EM) [16], impedance model (IM) [17], Schaaffs' collision factor theory (CFT) [18], Jacobson's free length theory (FLT) [19], and Prigogine–Flory–Patterson–Oswal theory (PFPOT) [20–23].

2. Experimental

All chemicals used in this study were of analytical grade obtained from s.d. fine-chem., Ltd. The claimed mass fraction purity for the chemicals was >0.995. These liquids were dried over 4 Å molecular sieves and partially degassed prior to use. The purity of these experimental liquids was checked by comparing the observed densities and speeds of sound with those reported in the literature. The measured values are presented in Table 1 along with the available literature values.

The densities of pure liquids and the binary mixtures were measured by calibrated single stem capillary pycnometer, having a bulb volume of about 10 cm^3 and capillary of internal diameter of 0.7 rnm. The speeds of sound in pure liquids and in their

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Table 1
Densities (ρ), speeds of sound (u), thermal expansion coefficients (α), and heat capacities (C_p^o) for pure liquids.

Liquid	T (K)	ρ (kg m^{-3})		u (m s^{-1})		α^{o} [a] (kK^{-1})	C_p^o (J mol^{-1} K^{-1})
		Expt.	Lit.	Expt.	Lit.		
DIPE	303.15	712.9	712.96[b] 713.06[d] 713.1[f] 713.20[g]	976	976.04[b] 974.9[e]	1.468	218[c]
	313.15	702.9	702.80[f] 702.59[g]	932	932.17[b]	1.489	222[h,i]
	323.15	691.9	691.533[b] 691.86[g]	889	888.61[b]	1.512	226[h,i]
Oxolane	303.15	876.9	877.0[j] 876.7[l]	1254	1254[j] 1255[l] 1256[m]	1.248	125[k]
	313.15	865.7	865.4[k] 870.5[o]	1207	1209[n] 1206.4[k]	1.265	128[k]
	323.15	854.8		1165		1.291	130[k]
DMA	303.15	948.4	948.0[p] 948.33[q]	1468		0.861	231[i]
	313.15	939.7	939.8[p] 939.72[q]	1431		0.868	235[i]
	323.15	931.5		1393		0.876	239[i]
DEA	303.15	926.1	925.9[p]	1434		0.862	291[i]
	313.15	917.5	918.0[p]	1396		0.869	296[i]
	323.15	964.6		1358		0.877	301[i]

[a] Derived from density data Ref. [24–30].
[b] Ref. [24].
[c] Ref. [31].
[d] Ref. [25].
[e] Ref. [32].
[f] Ref. [26].
[g] Ref. [27].
[h] Ref. [28].
[i] From group contribution method [33].
[j] Ref. [34].
[k] Ref. [28].
[l] Ref. [35].
[m] Ref. [36].
[n] Ref. [37].
[o] Ref. [38].
[p] Ref. [30].
[q] Ref. [39].

binary mixtures were measured using single-crystal variable-path ultrasonic interferometer (Mittal Enterprises, New Delhi, Model: M-82) operating at 2 MHz. For all the measurements, temperatures were controlled by circulating water through an ultra thermostat JULABO F-25 (made in Germany) keeping temperature fluctuations within ±0.02 K. The details of the experimental procedure have been described elsewhere [40]. The binary mixtures were prepared by mixing known masses of pure liquids in air tight stoppered bottles taking due precautions to minimize evaporation losses. All the mass measurements were performed on an electronic balance (Mettler-AE 240, Switzerland) accurate to ±0.05 mg. The samples for the measurement were immediately used after preparation. The uncertainty in density and speed of sound measurements was within 0.1 kg m^{-3} and 1 m s^{-1}.

3. Results and discussion

The results for the densities, speeds of sound, isentropic compressibilities, Rao's molar sound functions [41], specific acoustic impedances and intermolecular free lengths, excess molar volumes, and excess isentropic compressibilities for four binary mixtures of DIPE or oxolane with DMA or DEA at 303.15, 313.15, and 323.15 K are given in Tables 2–5.

The isentropic compressibilities κ_S, Rao's molar sound functions R, specific acoustic impedances Z and intermolecular free lengths L_f were obtained from the experimental densities ρ and speeds of sound u using the following relations:

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

$$R = u^{1/3} V \tag{2}$$

$$Z = u\rho \tag{3}$$

$$L_f = K_{\rm Jac}/(u\rho^{1/2}) = K_{\rm Jac}\kappa_S{}^{1/2} \tag{4}$$

where molar volume $V = \Sigma(x_i M_i)/\rho$, in which x_i and M_i are the mole fraction and molecular mass of component i. $K_{\rm Jac} = (91.368 + 0.3565T) \times 10^{-8}$ is temperature dependent Jacobson's constant [19]. The values of κ_S, R, and Z were uncertain within ±0.2% while L_f within 1%.

Excess molar volume V_m^E, excess isentropic compressibility κ_S^E, excess intermolecular free length L_f^E and excess specific acoustic impedance Z^E in each mixture were calculated with following expressions:

$$Y^E = Y - Y^{id} \tag{5}$$

where, Y represent either V, κ_S, L_f, and Z. The V_m^{id} for an ideal mixture was calculated from usual relation

$$V_m^{id} = \sum x_i V_i^o = \sum \frac{x_i M_i}{\rho_i} \tag{6}$$

Table 2
Densities (ρ), speeds of sound (u), isentropic compressibilities (κ_S), Rao's molar sound functions (R), specific acoustic impedances (Z) and intermolecular free lengths (L_f), excess molar volumes (V_m^E) and excess isentropic compressibilities (κ_S^E) for DIPE + DMA mixture at 303.15, 313.15 and 323.15 K.

x_1	ρ (kg m^{-3})	u (m s^{-1})	κ_S (TPa^{-1})	10^6R[a]	10^3Z (kg m^{-2} s^{-1})	$10L_f$ (nm)	$10^6\ V_m^E$ (m^3 mol^{-1})	κ_S^E (TPa^{-1})
303.15 K								
0	948.4	1468	489	1452	1392	0.439		
0.0741	932.6	1430	524	1447	1334	0.455	−0.498	−54
0.1589	914.0	1385	570	1441	1266	0.474	−0.966	−107
0.2428	895.3	1342	620	1436	1202	0.495	−1.351	−152
0.3355	874.3	1296	681	1432	1133	0.518	−1.681	−191
0.4294	852.6	1250	751	1428	1066	0.544	−1.892	−219
0.5338	827.8	1199	840	1425	993	0.576	−1.941	−232
0.6362	802.8	1149	944	1423	922	0.610	−1.780	−225
0.7466	775.2	1096	1074	1423	850	0.651	−1.366	−193
0.8676	744.8	1038	1246	1423	773	0.701	−0.701	−122
1	712.9	976	1473	1422	696	0.762		
313.15 K								
0	939.8	1431	520	1453	1345	0.461		
0.0741	924.8	1396	555	1448	1291	0.476	−0.654	−67
0.1589	906.5	1353	603	1442	1227	0.496	−1.217	−132
0.2428	887.5	1311	656	1438	1164	0.517	−1.608	−187
0.3355	866.2	1262	725	1432	1093	0.544	−1.942	−231
0.4294	844.2	1214	804	1428	1025	0.573	−2.151	−263
0.5338	819.2	1161	906	1425	951	0.608	−2.210	−279
0.6362	794.1	1112	1018	1423	883	0.645	−2.067	−275
0.7466	766.4	1059	1164	1423	812	0.689	−1.661	−242
0.8676	735.7	1000	1359	1423	736	0.745	−0.958	−162
1	702.5	932	1639	1421	655	0.818		
323.15 K								
0	931.5	1393	553	1453	1298	0.484		
0.0741	916.9	1360	590	1447	1247	0.500	−0.767	−81
0.1589	898.6	1318	641	1442	1184	0.521	−1.391	−159
0.2428	879.6	1277	697	1438	1123	0.544	−1.839	−225
0.3355	858.3	1229	771	1433	1055	0.572	−2.233	−281
0.4294	836.3	1182	856	1429	989	0.602	−2.499	−322
0.5338	811.5	1128	969	1424	915	0.641	−2.649	−343
0.6362	786.4	1078	1094	1422	848	0.681	−2.557	−342
0.7466	758.5	1024	1257	1422	777	0.730	−2.162	−306
0.8676	726.9	961	1490	1421	699	0.795	−1.321	−205
1	691.9	889	1829	1420	615	0.880		

[a] Unit = m$^{10/3}$ s$^{-1/3}$ mol^{-1}.

κ_S^{id} for an ideal mixture were calculated from the relation recommended by Benson and Kiyohara [42]

$$\kappa_S^{id} = \sum \phi_i \left\{ \frac{\kappa_{S,i}^{o} + TV_i^{o}(\alpha_i^{o})^2}{C_{p,i}^{o}} \right\} - T\left(\sum x_i V_i^{o}\right) \frac{\sum \phi_i \alpha_i^{o\,2}}{\sum x_i C_{p,i}^{o}} \tag{7}$$

L_f^{id} and Z^{id} for an ideal mixture were obtained as

$$L_f^{id} = K_{Jac}(\kappa_S^{id})^{1/2} \tag{8}$$

$$Z^{id} = \rho^{id} u^{id} = \left(\frac{\rho^{id}}{\kappa_S^{id}}\right)^{1/2} \tag{9}$$

in which the M_i, V_i^o, α_i^o and $C_{p,i}^o$ are respectively, the molar mass, molar volume, isobaric thermal expansion coefficient and molar isobaric heat capacity of pure component i, $\phi_i = x_iV_i/\Sigma x_jV_j$ is the volume fraction of i in the mixture, stated in terms of the unmixed components and $\rho^{id} = \sum \phi_i \rho_i^o$. The values of α^o and C_p^o used for these calculations are included in Table 1.

For compact and smooth representation, the values of u and κ_S were fitted to a polynomial equation of the form

$$u \text{ or } \kappa_S \text{ or } R = \sum_{i=0}^{m} A_i X_1^i \tag{10}$$

The V_m^E, κ_S^E, L_f^E and Z^E were correlated by Redlich–Kister polynomial equation

$$Y^E = x_1(1-x_1)\sum_{i=0}^{n} B_i(1-2x_1)^i \tag{11}$$

where $Y^E = V_m^E,\ \ \kappa_S^E,\ \ L_f^E$, and Z^E.

The coefficients A_i of Eq. (10) and B_i of Eq. (11) and corresponding standard deviations σ obtained from a least-squares fit with equal weights assigned to each point are given in Tables 6 and 7.

It is observed from Tables 2–5, that the values of speeds of sound, specific acoustic impedances decrease with mole fraction of DIPE or oxolane and also with the rise in temperature for all the mixtures studied. The values of isentropic compressibility and intermolecular free length increase with the mole fraction of DIPE or oxolane and with the rise in temperature from 303.15 to 323.15 K. The Figs. 1–4 show the dependence of V_m^E, κ_S^E, L_f^E and Z^E on composition. The V_m^E, κ_S^E and L_f^E are negative for all the four studied mixtures over the entire mole fraction range, and their magnitude increases with the rise in temperature from 303.15 to 323.15 K. Further, the values of V_m^E for both the DIPE mixtures are much more negative compared to both oxolane mixtures. Other very large negative values of V_m^E reported in the literature include 2-methoxyethanol + tert-butylamine (−2.4 to −2.6 × 10^{-6} m^3 mol^{-1} [43]) and DMSO + cyclohexylamine (−3.4 × 10^{-6} m^3 mol^{-1}) [44].

The observed values of V_m^E for presently investigated DIPE + DMA and +DEA mixtures are less negative than DIPE + aniline [7], +toluidines [8]. Similarly, V_m^E for presently investigated oxolane + DMA

Table 3

Densities (ρ), speeds of sound (u), isentropic compressibilities (κ_S), Rao's molar sound functions (R), specific acoustic impedances (Z) and intermolecular free lengths (L_f), excess molar volumes (V_m^E) and excess isentropic compressibilities (κ_S^E) for DIPE + DEA mixture at 303.15, 313.15 and 323.15 K.

x_1	ρ (kg m^{-3})	u (m s^{-1})	κ_S (T Pa^{-1})	$10^6 R$[a]	$10^3 Z$ (kg m^{-2} s^{-1})	$10 L_f$ (nm)	$10^6 V_m^E$ (m^3 mol^{-1})	κ_S^E (T Pa^{-1})
303.15 K								
0	926.2	1434	525	1817	1328	0.455		
0.078	914.4	1403	556	1782	1283	0.468	−0.546	−43
0.1606	901.0	1370	591	1746	1234	0.483	−1.023	−86
0.2581	884.0	1330	640	1705	1176	0.502	−1.453	−130
0.3421	868.5	1295	687	1671	1125	0.520	−1.742	−163
0.4336	850.7	1255	746	1634	1068	0.543	−1.967	−191
0.5341	829.8	1209	825	1593	1003	0.570	−2.063	−209
0.6457	804.7	1156	930	1550	930	0.606	−1.938	−210
0.7545	778.3	1103	1056	1510	859	0.645	−1.570	−187
0.8762	746.7	1042	1233	1466	778	0.698	−0.889	−125
1	712.9	976	1473	1422	696	0.762		
313.15 K								
0	917.6	1396	559	1818	1281	0.478		
0.078	906.8	1367	590	1782	1240	0.491	−0.770	−53
0.1606	893.6	1336	627	1746	1194	0.506	−1.329	−106
0.2581	876.7	1296	679	1705	1136	0.527	−1.830	−160
0.3421	861.1	1260	732	1670	1085	0.547	−2.144	−199
0.4336	843.0	1218	800	1632	1027	0.571	−2.359	−230
0.5341	822.1	1171	887	1591	963	0.602	−2.499	−252
0.6457	797.1	1117	1006	1547	890	0.641	−2.435	−255
0.7545	770.6	1063	1148	1506	819	0.685	−2.081	−230
0.8762	738.3	1000	1355	1463	738	0.744	−1.290	−154
1	702.5	932	1639	1421	655	0.818		
323.15 K								
0	909.2	1358	596	1818	1235	0.503		
0.078	899.0	1332	627	1782	1198	0.516	−0.936	−66
0.1606	886.6	1302	665	1745	1154	0.531	−1.696	−130
0.2581	870.2	1261	723	1702	1097	0.553	−2.352	−194
0.3421	854.7	1224	781	1666	1046	0.575	−2.737	−240
0.4336	836.7	1182	856	1628	989	0.602	−3.026	−279
0.5341	815.7	1134	953	1587	925	0.636	−3.205	−307
0.6457	790.6	1080	1084	1542	854	0.678	−3.180	−314
0.7545	764.0	1023	1251	1500	782	0.728	−2.857	−282
0.8762	730.8	958	1491	1457	700	0.795	−1.927	−190
1	691.9	889	1829	1420	615	0.880		

[a] Unit = m$^{10/3}$ s$^{-1/3}$ mol^{-1}.

and +DEA mixtures are less negative than oxolane + aniline [6] +monoalkylaniline [6], +toluidines [8]. This clearly indicates influence of tertiary amine over primary and secondary amines.

The over all behaviour of V_m^E and κ_S^E for the present mixtures can be envisaged as a resultant of opposite effects: (i) disruption of ether–ether interactions; (ii) break down of dipolar order in ether and tertiary arylamine; (iii) dipole–dipole interactions in tertiary aryl amine and ether; (iv) free volume effect; and (v) characteristic pressure P^* effect [45,46]. The free volume (V_{fs}^E), characteristic pressure (V_{P*}^E) and interaction (V_{int}^E) contributions to the total V_{total}^E can be estimated from the Prigogine–Flory–Patterson (PFP) theory [26–28]. Table 8 summarizes these contributions at equimolar composition for the present mixtures. The estimated sum of free volume and P^* contributions ($V_{fv}^E + V_{P*}^E$) to the V_{total}^E as per the PFP theory is in the range from −1.821 to -2.291×10^{-6} m^3 mol^{-1} DIPE + DMA or DEA and −0.152 to -0.296×10^{-6} m^3 mol^{-1} oxolane + DMA or DEA, while interaction contribution is in the range from −0.114 to -0.968×10^{-6} m^3 mol^{-1} DIPE + DMA or DEA and −0.213 to -0.536×10^{-6} m^3 mol^{-1} oxolane + DMA or DEA. This points to the fact that in very large negative values of V_m^E for DIPE + DMA or DEA the major contribution is because of the free volume and P^* effects, which include size, shape and conformation of the molecules involved. In case of oxolane mixtures, the combined contribution due to the free volume and P^* effects is much smaller as compared to the DIPE mixtures. This difference can be explained by considering the difference in thermal expansion coefficient α or Flory's reduced volume $\tilde{V}$ and characteristic pressure P^* of unlike components (listed in Tables 1 and 9). The free volume (V_{fv}^E) term is proportional to $-(\tilde{V}_1 - \tilde{V}_2)^2$ i.e. $-(\alpha_1 - \alpha_2)^2$ [22,45] and is always negative and its magnitude depends upon difference in thermal expansion coefficients or Flory's reduced volumes of involved components. The difference between α or $\tilde{V}$ of DIPE and DMA/DEA is relatively very large as compared to the α or $\tilde{V}$ of oxolane and DMA/DEA, that is the reason free volume (V_{fv}^E) term for DIPE + DMA/MEA is very large negative compared to the oxolane + DMA/DEA mixtures. The characteristic pressure (V_{P*}^E) term is proportional to $(P_1^* - P_2^*)(\tilde{V}_1 - \tilde{V}_2)$ and can have either sign depending upon the magnitude of P^* and α (or $\tilde{V}$) of unlike components [22,45]. For DIPE + DMA/MEA the $(P_1^* - P_2^*)$ is large negative while $(\tilde{V}_1 - \tilde{V}_2)$ large positive, making (V_{P*}^E) term large and negative. For oxolane + DMA, $(P_1^* - P_2^*)$ is small negative and $(\tilde{V}_1 - \tilde{V}_2)$ positive, with product $(P_1^* - P_2^*)(\tilde{V}_1 - \tilde{V}_2)$ i.e. (V_{P*}^E) term for oxolane + DMA is small negative. Contrary to this for oxolane + DEA, $(P_1^* - P_2^*)$ is small positive and $(\tilde{V}_1 - \tilde{V}_2)$ positive, as a result V_{P*}^E is small and positive.

The negative values of interactional contribution (V_{int}^E) to V_m^E for the present mixtures suggest that dipole–dipole interactions (μ for DIPE/oxolane = 1.34/1.75 D, and for DMA/DEA ≈1.61 D), between unlike molecules dominate over the dispersive ether–ether interactions, breaking of dipolar order of aromatic amines as well as of DIPE/oxolane. The negative V_m^E and κ_S^E values at equimolar compo-

Table 4

Densities (ρ), speeds of sound (u), isentropic compressibilities (κ_S), Rao's molar sound functions (R), specific acoustic impedances (Z) and intermolecular free lengths (L_f), excess molar volumes (V_m^E) and excess isentropic compressibilities (κ_S^E) for oxolane + DMA mixture at 303.15, 313.15 and 323.15 K.

x_1	ρ (kg m^{-3})	u (m s^{-1})	κ_S (T Pa^{-1})	10^6 R[a]	$10^3 Z$ (kg m^{-2} s^{-1})	$10 L_f$ (nm m^3)	10^6 V_m^E (mol^{-1})	κ_S^E (T Pa^{-1})
303.15 K								
0	948.4	1468	489	1452	1392	0.439		
0.0683	945.8	1459	497	1413	1380	0.443	−0.082	−6
0.1883	940.8	1439	513	1343	1354	0.450	−0.214	−13
0.2797	936.7	1423	527	1290	1333	0.456	−0.319	−19
0.389	931.0	1403	546	1228	1306	0.464	−0.400	−24
0.4723	926.2	1387	561	1180	1285	0.471	−0.451	−28
0.5757	919.3	1365	584	1121	1255	0.480	−0.467	−31
0.6813	910.9	1340	611	1062	1221	0.491	−0.415	−30
0.7866	901.1	1314	643	1004	1184	0.504	−0.306	−26
0.8909	890.1	1287	678	947	1146	0.517	−0.173	−18
1	876.9	1254	725	887	1100	0.535		
313.15 K								
0	939.8	1431	520	1453	1345	0.461		
0.0683	937.5	1424	526	1414	1335	0.464	−0.141	−9
0.1883	932.9	1408	541	1345	1314	0.470	−0.356	−22
0.2797	928.6	1391	557	1292	1292	0.477	−0.458	−28
0.3890	922.9	1370	577	1229	1264	0.486	−0.565	−35
0.4723	918.0	1354	594	1181	1243	0.493	−0.625	−41
0.5757	911.1	1331	620	1122	1213	0.503	−0.666	−44
0.6813	902.8	1304	651	1062	1177	0.516	−0.648	−43
0.7866	893.2	1276	688	1003	1140	0.530	−0.582	−39
0.8909	881.4	1246	731	946	1098	0.546	−0.390	−28
1	865.7	1207	793	887	1045	0.569		
323.15 K								
0	931.5	1393	553	1453	1298	0.484		
0.0683	929.5	1391	556	1415	1293	0.485	−0.202	−14
0.1883	925.2	1376	571	1346	1273	0.492	−0.490	−30
0.2797	921.1	1359	588	1292	1252	0.499	−0.640	−38
0.389	915.3	1337	611	1229	1224	0.509	−0.762	−46
0.4723	910.2	1319	632	1181	1201	0.517	−0.818	−51
0.5757	903.0	1294	661	1121	1169	0.529	−0.850	−54
0.6813	894.4	1267	697	1062	1133	0.543	−0.825	−54
0.7866	884.2	1238	738	1003	1095	0.559	−0.721	−48
0.8909	871.7	1206	789	946	1051	0.578	−0.483	−34
1	854.8	1165	862	888	996	0.604		

[a] Unit=m$^{10/3}$ s$^{-1/3}$ mol$^{-1.}$

Table 5

Densities (ρ), speeds of sound (u), isentropic compressibilities (κ_S), Rao's molar sound functions (R), specific acoustic impedances (Z) and intermolecular free lengths (L_f), excess molar volumes (V_m^E) and excess isentropic compressibilities (κ_S^E) for oxolane + DEA mixture at 303.15, 313.15 and 323.15 K.

x_1	ρ (kg m^{-3})	u (m s^{-1})	κ_S (T Pa^{-1})	10^6 R[a]	$10^3 Z$ (kg m^{--2} s^{-1})	$10 L_f$ nm	10^6 V_m^E (m^3 mol^{-1})	κ_S^E (T Pa^{-1})
303.15 K								
0	0.9262	1434	525	1817	1328	0.455		
0.0933	0.9245	1428	530	1730	1320	0.457	−0.126	−7
0.1907	0.9222	1418	539	1639	1308	0.461	−0.205	−13
0.2899	0.9196	1405	551	1545	1292	0.466	−0.286	−17
0.3879	0.9165	1390	565	1453	1274	0.472	−0.334	−20
0.4892	0.9128	1372	582	1357	1252	0.479	−0.374	−22
0.5937	0.9080	1352	603	1260	1228	0.488	−0.360	−23
0.6904	0.9029	1331	625	1169	1202	0.497	−0.347	−22
0.7948	0.8960	1307	653	1073	1171	0.508	−0.279	−19
0.8935	0.8879	1283	684	983	1139	0.520	−0.170	−13
1	0.8769	1254	725	887	1100	0.535		
313.15 K								
0	0.9176	1396	559	1818	1281	0.478		
0.0933	0.9158	1390	565	1731	1273	0.480	−0.135	−9
0.1907	0.9136	1379	576	1639	1260	0.485	−0.257	−15
0.2899	0.9109	1366	588	1546	1244	0.490	−0.349	−20
0.3879	0.9077	1351	604	1453	1226	0.496	−0.409	−25
0.4892	0.9039	1333	623	1358	1205	0.504	−0.463	−28
0.5937	0.8992	1312	646	1259	1180	0.514	−0.490	−29
0.6904	0.8938	1291	671	1169	1154	0.524	−0.467	−29
0.7948	0.8868	1265	705	1072	1122	0.536	−0.416	−25
0.8935	0.8781	1240	741	983	1089	0.550	−0.272	−19
1	0.8657	1207	793	887	1045	0.569		
323.15 K								
0	0.9092	1358	596	1818	1235	0.503		
0.0933	0.9076	1354	601	1731	1229	0.505	−0.195	−12
0.1907	0.9055	1343	612	1639	1216	0.509	−0.358	−19

Table 5 (*Continued*)

x_1	ρ (kg m^{-3})	u (m s^{-1})	κ_S (T Pa^{-1})	10^6 R[a]	$10^3 Z$ (kg m^{--2} s^{-1})	$10 L_f$ nm	10^6 V_m^E (m^3 mol^{-1})	κ_S^E (T Pa^{-1})
0.2899	0.9028	1330	626	1546	1201	0.515	−0.474	−26
0.3879	0.8995	1315	643	1453	1183	0.522	−0.543	−31
0.4892	0.8953	1296	665	1358	1160	0.531	−0.565	−34
0.5937	0.8904	1275	691	1260	1135	0.541	−0.594	−36
0.6904	0.8849	1252	721	1169	1108	0.553	−0.586	−35
0.7948	0.8778	1226	758	1072	1076	0.567	−0.552	−31
0.8935	0.8687	1200	799	983	1042	0.582	−0.391	−24
1	0.8548	1165	862	888	996	0.604		

[a] Unit = m$^{10/3}$ s$^{-1/3}$ mol^{-1}.

sitions for common amine follow the order: DIPE > oxolane, while there is a marginal difference between the values of DMA or DEA.

As expected, the trend of negative L_f^E values (Fig. 3 and Table 7) is similar to κ_S^E in the studied mixtures. The negative values of L_f^E are generally observed in the mixtures with specific interactions between unlike molecules. The negative L_f^E values at equimolar compositions for common amine follow the order: DIPE > oxolane. Fig. 4 shows that Z^E is positive for all the studied mixtures. Positive trends in Z^E have also been reported for binary mixtures of methanol + piperidine [47]. Specific acoustic impedance is a quantity, which depends on the molecular packing of the systems. The positive values of Z^E are in accordance with the presence of struc-

Table 6
Coefficients A_i of Eq. (10) along with standard deviations σ of binary mixture properties.

Properties	T (K)	A_0	A_1	A_2	A_3	A_4	σ (Y^E)
DIPE + DMA							
u (m s^{-1})	303.15	1468.2	−529.4	55.1	−18.2		0.5
	313.15	1430.9	−454	−275.3	461.9	−231.5	0.5
	323.15	1393.1	−433.7	−284.3	435.4	−221.7	0.7
κ_S (T Pa^{-1})	303.15	488.8	475.3	209.9	197.1	101.8	0.6
	313.15	520.4	394.7	833.8	−780.0	669.5	1.2
	323.15	554.1	399.1	1003.5	−1072.4	944	1.7
$10^6 R$[a]	303.15	1452.7	−87.6	82.5	−25.2		0.6
	313.15	1453.5	−87.9	79.8	−24.0		0.9
	323.15	1452.7	−73.3	41.4			0.7
DIPE + DEA							
u (m s^{-1})	303.15	1433.6	−382.6	−67.1	−7.8		0.5
	313.15	1395.6	−340.9	−212.7	155.4	−65.3	0.5
	323.15	1358.1	−310.0	−313.9	262	−107.5	0.6
κ_S (T Pa^{-1})	303.15	525.5	348.2	385.2	−201.7	414.7	1.1
	313.15	560.3	311.2	706.6	−652.5	712.5	1.5
	323.15	597.3	293.2	944.2	−990.5	984.1	1.3
$10^6 R$[a]	303.15	1816.7	−444.6	50.0			0.4
	313.15	1816.9	−441.9	24.2	21.4		0.6
	323.15	1816.9	−444.6	−5.6	52.3		1.0
Oxolane + DMA							
u (m s^{-1})	303.15	1468.5	−147.5	−43.6	−23.1		0.6
	313.15	1431.0	−85.8	−257.5	256.3	−136.9	0.8
	323.15	1393.4	−13.7	−523.7	591.5	−282.4	0.6
κ_S (T Pa^{-1})	303.15	489.1	111.0	90.2	−31.7	66.4	0.5
	313.15	519.8	66.5	304.0	−344.9	247.1	0.9
	323.15	553.2	6.6	573.4	−690.8	419.2	0.7
$10^6 R$[a]	303.15	1452.8	−587.5	21.4			0.3
	313.15	1453.1	−572.8	−20.2	26.7		0.2
	323.15	1453.4	−568.0	−38.5	40.9		0.4
Oxolane + DEA							
u (m s^{-1})	303.15	1434.4	−60.4	−150.8	31.1		0.5
	313.15	1396.1	−51.4	−217.6	154.8	−74.8	0.5
	323.15	1358.2	−28.3	−295.1	244.9	−114.5	0.8
κ_S (T Pa^{-1})	303.15	525.0	41.1	195.7	−131.4	94.6	0.2
	313.15	559.3	43.4	247.0	−224.1	167.0	0.7
	323.15	596.5	19.2	378.8	−395.4	262.4	1.1
$10^6 R$[a]	303.15	1817.1	−929.6	−38.0	37.4		0.2
	313.15	1817.8	−930.0	−42.0	41.1		0.2
	323.15	1817.8	−927.0	−51.4	48.0		0.4

[a]Unit = (m$^{10/3}$ s$^{-1/3}$ mol^{-1}).

Table 7
Coefficients B_i of Eq. (11) along with standard deviations $\sigma\ (Y^E)$ of excess properties of binary mixtures.

Properties	T (K)	B_0	B_1	B_2	B_3	$\sigma\ (Y^E)$
DIPE + DMA						
$10^6\ V_m^E (m^3\ mol^{-1})$	303.15	−7.798	0.341	2.094	−2.045	0.002
	313.15	−8.832	0.525	0.144	−2.043	0.007
	323.15	−10.470	2.120	−1.066	−2.801	0.006
$\kappa_S^E\ (TPa^{-1})$	303.15	−917.9	201.8	−27.7	−40.6	0.4
	313.15	−1105.1	252.1	−169.4	61.2	0.6
	323.15	−1360.2	380.0	−233.6	56.0	1.3
$10^3 Z^E\ (kg\ m^{-2}\ s^{-1})$	303.15	493.3	356.5	202.9		3.3
	313.15	521.1	390.0	285.4		3.3
	323.15	561.9	411.8	315.3		4.1
$10\ L_f^E$ (nm)	303.15	−0.304	−0.023			0.001
	313.15	−0.368	−0.021			0.003
	323.15	−0.442	−0.011			0.003
DIPE + DEA						
$10^6\ V_m^E\ (m^3\ mol^{-1})$	303.15	−8.182	1.272	0.549	−1.494	0.006
	313.15	−9.880	2.146	−2.073	−1.853	0.010
	323.15	−12.650	2.868	−4.617	0.819	0.005
$\kappa_S^E (TPa^{-1})$	303.15	−815.9	317.1	−117.7	57.9	0.6
	313.15	−986.5	381.6	−187.2	96.0	0.2
	323.15	−1198.8	489.4	−254.1	84.6	0.9
$10^3 Z^E\ (kg\ m^{-2}\ s^{-1})$	303.15	465.6	192.7	69.7		0.6
	313.15	495.7	223.7	110.7		0.8
	323.15	530.3	253.7	145.8		1.5
$10\ L_f^E$ (nm)	303.15	−0.276	0.042			0.001
	313.15	−0.329	0.046			0.002
	323.15	−0.393	0.052			0.002
Oxolane + DMA						
$10^6 V_m^E\ (m^3\ mol^{-1})$	303.15	−1.823	0.443	0.462	−0.276	0.007
	313.15	−2.547	0.876	−0.960	0.423	0.007
	323.15	−3.330	0.758	−1.228	0.632	0.002
$\kappa_S^E (TPa^{-1})$	303.15	−113.2	60.0	−35.5		0.6
	313.15	−163.2	90.4	−74.0		1.1
	323.15	−204.1	74.6	−125.0	20.9	0.8
$10^3 Z^E\ (kg\ m^{-2}\ s^{-1})$	303.15	138.3	−23.5			0.8
	313.15	174.0	−32.2	50.2		0.8
	323.15	197.4	−2.3	93.4		0.7
$10\ L_f^E$ (nm)	303.15	−0.045	0.019			0.001
	313.15	−0.073	0.026			0.001
	323.15	−0.094	0.019			0.001
Oxolane + DEA						
$10^6\ V_m^E\ (m^3\ mol^{-1})$	303.15	−1.462	0.362	0.248	−0.242	0.008
	313.15	−1.868	0.449	−0.607	0.245	0.006
	323.15	−2.310	0.478	−1.477	1.093	0.007
$\kappa_S^E (TPa^{-1})$	303.15	−87.6	30.3	−36.7		0.4
	313.15	−108.3	41.1	−60.7	29.8	0.7
	323.15	−133.6	43.7	−91.4	44.2	1.2
$10^3 Z^E\ (kg\ m^{-2}\ s^{-1})$	303.15	95.3	−3.1			1.2
	313.15	106.1	−22.4	44.9		0.7
	323.15	122.8	−20.9	70.1		1.0
$10\ L_f^E$ (nm)	303.15	−0.037	0.007			0.001
	313.15	−0.048	0.016			0.001
	323.15	−0.062	0.019			0.001

tural effect and specific interactions between the unlike molecules [6–8].

The dependence of Rao's molar sound functions R on mole fractions x_1 is shown in Fig. 5. It is observed that there are slight negative deviations from the linear dependence on x_1 for DIPE binary mixtures while almost linear for oxolane mixtures. As expected the R are independent of temperature (Tables 2–5). According to Rao's approach [41], the speed of sound is directly related to the strength of intermolecular energy. If the intermolecular energy were simply a consequence of additive non-bonding interaction, the Rao's molar sound function should predict additive behaviour. The fact that is not so implies some additional interactions in the mixtures compared with those in the simple liquids [48].

4. Estimation of speeds of sound

Recently Gayol et al. [49] and Khammer and Shaw [50] described and tested several predictive methods for speed of sound in alkanol + n-alkanes mixtures and discussed the combing rules used in different models. Glinski [51] discussed the additivity of sound velocity in twenty four randomly selected binary mixtures and found Nomoto model [14] based on Rao's hypothesis [41] provides

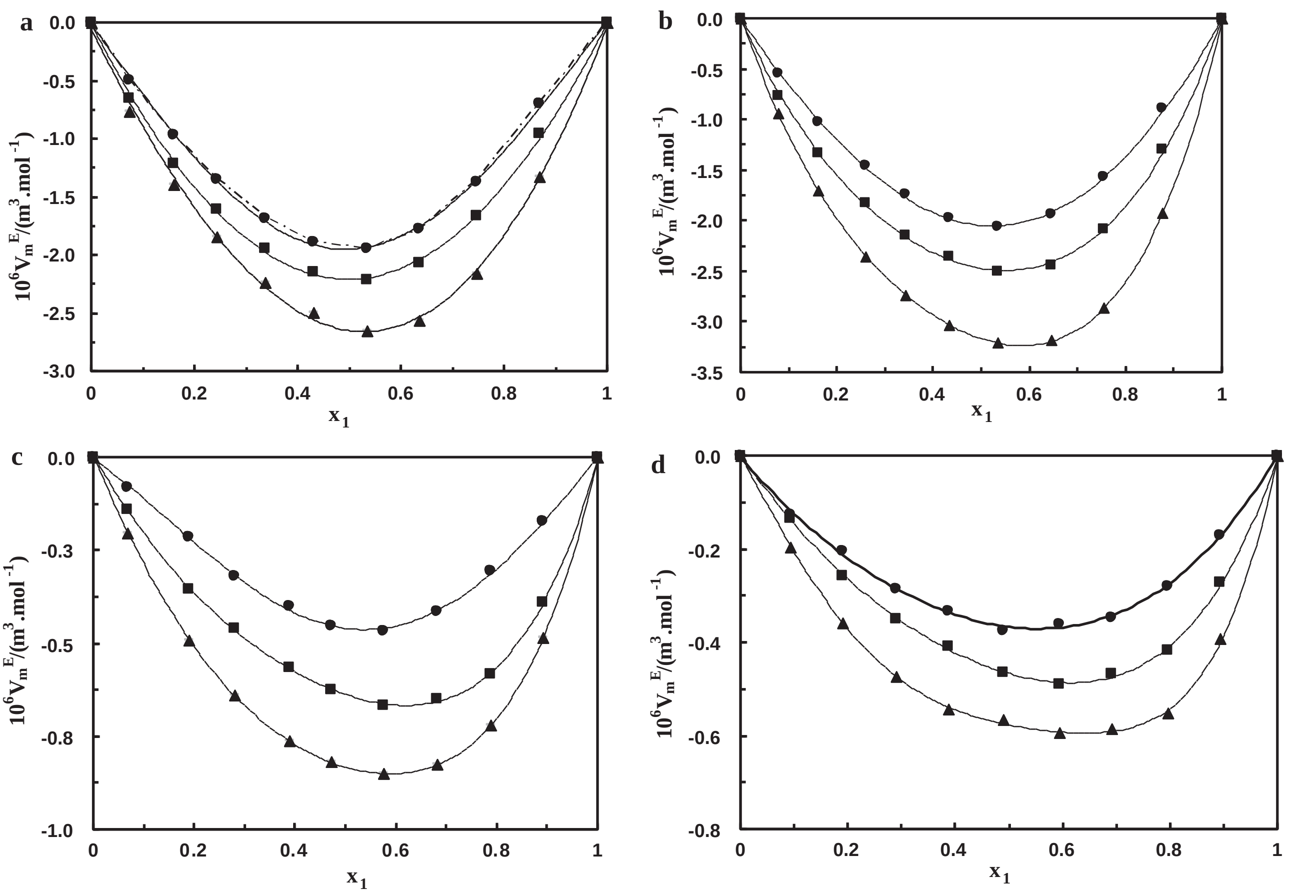

Fig. 1. Dependence of excess molar volumes on mole fraction of DIPE or oxolane, (a) DIPE + DMA, (b) DIPE + DEA, (c) oxolane + DMA and (d) oxolane + DEA at T = 303.15 K (●), 313.15 K (■), and 323.15 K (▲). Solid lines have been drawn from Eq. (11) using coefficient given in Table 7.

results similar to those of Ernst et al. model [16] while the Van Dael model [15] often fails. For all three models large deviations were observed for mixtures with associating constituents. Here, the speeds of sound in the investigated binary mixtures have been estimated at 303.15, 313.15 and 323.15 K from the Nomoto model [14] Van Dael model [15], Ernst et al. model [16], impedance model [17], collision factor theory [18], intermolecular free length theory [19], and Prigogine–Flory–Patterson–Oswal theory [20–23]. The pertinent relations in these calculations and their theoretical basis have been outlined several times and will not be repeated here. PFPO model is the only model amongst the considered here which has interaction parameter X_{12}. Normally X_{12} is obtained from excess molar enthalpy data [21]. In absence of excess molar enthalpy data needed to determine interaction parameter X_{12}, we have determined speed of sound using $X_{12} = 0$ as well as X_{12} derived from equation [21]

$$X_{12} = P_1^*\left[1-\left(\frac{P_2^*}{P_1^*}\right)^{1/2}\left(\frac{V_2^*}{V_1^*}\right)^{1/6}\right]^2 \tag{12}$$

Table 8

Free volume contribution $V_{\mathrm{fv}}^{\mathrm{E}}$, P^* contribution V_{P*}^{E} and interaction contribution to $V_{\mathrm{int}}^{\mathrm{E}}$ to total excess molar volume $V_{\mathrm{total}}^{\mathrm{E}}$ at equimolar composition as per the PFP theory.

T (K)	$10^6\ V_{\mathrm{fv}}^{\mathrm{E}}$ (m³ mol⁻¹)	$10^6\ V_{P*}^{\mathrm{E}}$ (m³ mol⁻¹)	$10^6\ (V_{\mathrm{fv}}^{\mathrm{E}}+V_{P*}^{\mathrm{E}})$ (m³ mol⁻¹)	$10^6\ V_{\mathrm{int}}^{\mathrm{E}}$ (m³ mol⁻¹)	$10^6\ V_{\mathrm{total}}^{\mathrm{E}}$ (m³ mol⁻¹)
DIPE + DMA					
303.15	−0.558	−1.288	−1.846	−0.101	−1.947
313.15	−0.603	−1.417	−2.020	−0.188	−2.208
323.15	−0.681	−1.610	−2.291	−0.327	−2.618
DIPE + DEA					
303.15	−0.597	−1.224	−1.821	−0.224	−2.045
313.15	−0.643	−1.344	−1.987	−0.483	−2.470
323.15	−0.692	−1.500	−2.192	−0.970	−3.162
THF + DMA					
303.15	−0.181	−0.042	−0.223	−0.233	−0.456
313.15	−0.201	−0.053	−0.254	−0.383	−0.637
323.15	−0.236	−0.060	−0.296	−0.536	−0.832
THF + DEA					
303.15	−0.199	0.047	−0.152	−0.213	−0.365
313.15	−0.221	0.047	−0.174	−0.283	−0.457
323.15	−0.243	0.050	−0.193	−0.385	−0.578

Table 9
Flory's characteristic and reduced parameters of neat components.

Component	T (K)	$\tilde{V}$	10^6V^* (m^3 mol^{-1})	$10^{-6}P^*$ (J m^{-3})
DIPE	303.15	1.3407	106.85	419
	313.15	1.3529	107.45	408
	323.15	1.3655	108.15	394
Oxolane	303.15	1.3004	63.21	617
	313.15	1.3122	63.45	612
	323.15	1.3242	63.67	608
DMA	303.15	1.2216	104.60	634
	313.15	1.2293	104.90	632
	323.15	1.2343	105.40	629
DEA	303.15	1.2218	131.88	600
	313.15	1.2295	132.28	596
	323.15	1.2375	132.64	592

where P^* and V^* are Flory's reduction parameters [21]. In order to determine relative predicting ability of various models, the standard percentage deviation $\sigma\%$ was calculated by considering percentage deviations for all mole fractions using the relation (13) and recorded in Table 10.

$$\sigma\% \left[\sum \left\{\frac{100(u_{\exp}-u_{\mathrm{cal}})}{u_{\exp}}\right\}^{2} /n\right]^{1/2} \qquad (13)$$

where n represents number of data points.

The study of Table 10 reveals that the values of $\sigma\%$ for the NM, VM, EM, IM, CFT, FLT, and PFPOT are in the range from 0.1 to 2.8, 5.7 to 10.6, 0.3 to 5.2, 0.4 to 2.7, 0.4 to 1.3, 0.6 to 3.3 and 0.2 to 1.7 (0.1–1.2), respectively for the four binary mixtures at investigated three different temperatures. The average values of standard deviations $\langle\sigma\%\rangle$ 1.2%, 1.5%, 0.9%, 1.5%, and 0.9% for the NM, IM, CFT, FLT and PFPOT, respectively are comparatively less than those found for other estimations. In EM and VM average $\sigma\%$ are 2.3 and 7.9%. Further it is observed, the estimated results by all the models in oxolane mixtures are better than those for the mixtures involving DIPE. This is attributed to the

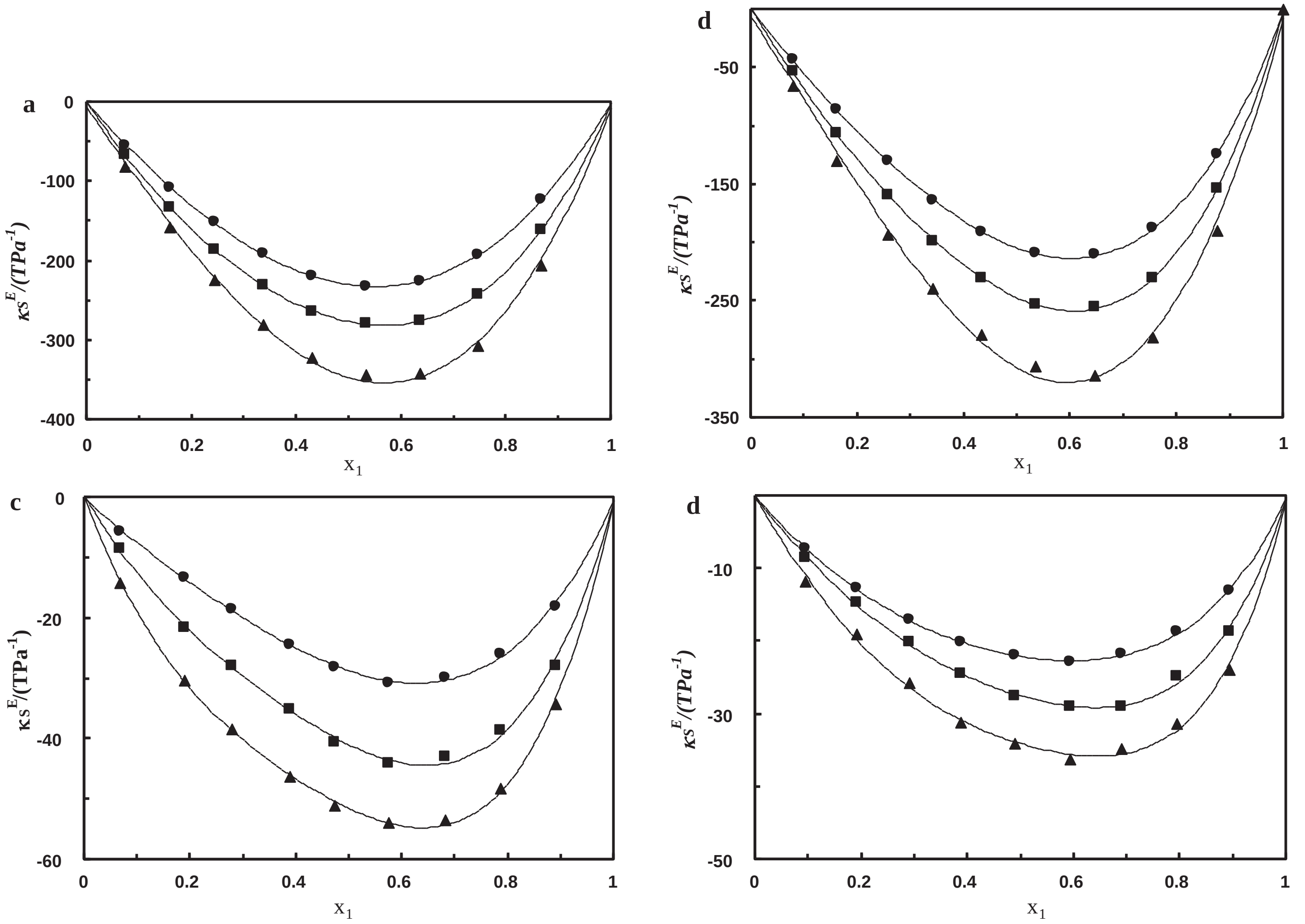

Fig. 2. Dependence of excess isentropic compressibilities on mole fraction of DIPE or oxolane, (a) DIPE + DMA, (b) DIPE + DEA, (c) oxolane + DMA and (d) oxolane + DEA at T = 303.15 K (●), 313.15 K (■), 323.15 K (▲). Solid lines have been drawn from Eq. (11) using coefficient given in Table 7.

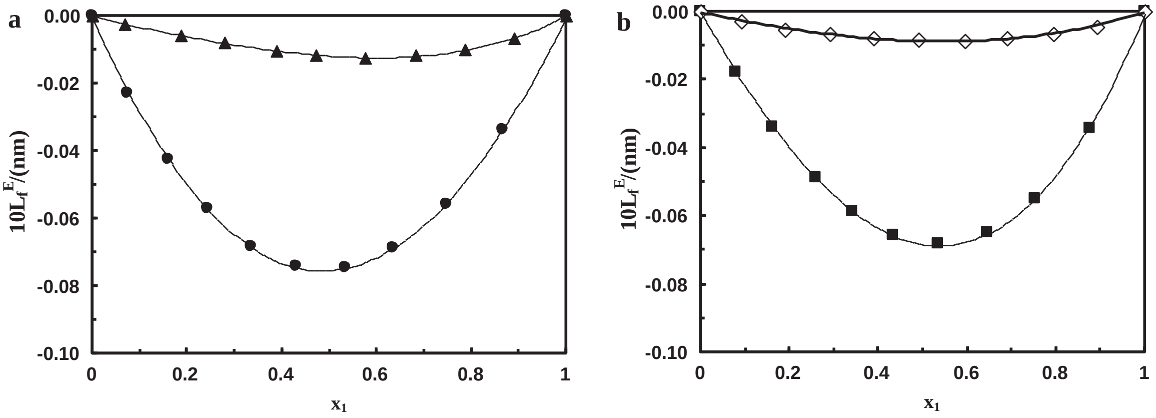

Fig. 3. Dependence of excess intermolecular free length on mole fraction of DIPE or oxolane at T = 303.15 K. (a) DIPE + DMA (●) and oxolane + DMA (▲). (b) DIPE + DEA (■) and oxolane + DEA (◊), Solid lines have been drawn from Eq. (11) using coefficient given in Table 7.

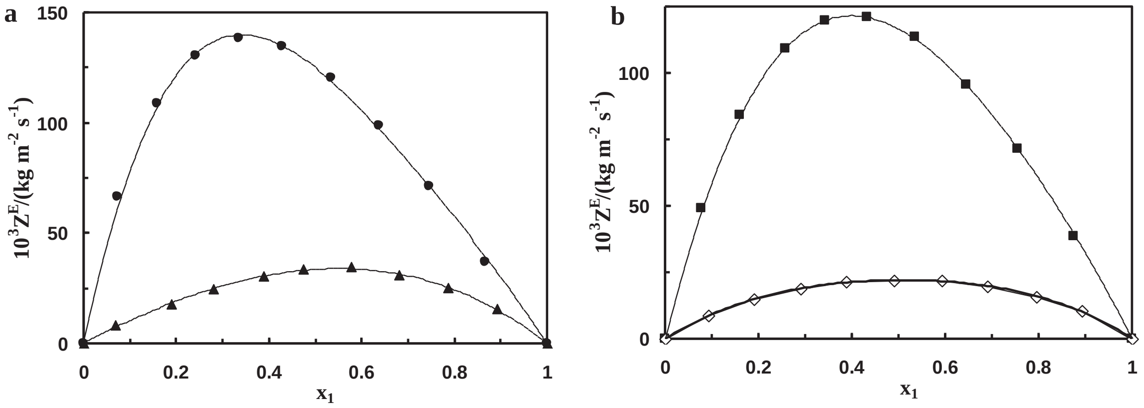

Fig. 4. Dependence of excess specific acoustic impedance on mole fraction of DIPE or oxolane at T = 303.15 K. (a) DIPE + DMA (●) and oxolane + DMA (▲). (b) DIPE + DEA (■) and oxolane + DEA (◊), Solid lines have been drawn from Eq. (11) using coefficient given in Table 7.

fact that in oxolane mixtures, the interactions are not so complex as in the case of DIPE mixtures. Considering the average values of standard deviations, the estimation ability of speed of sound in the presently investigated mixtures follows the sequence PFPOT > CFT > NM > FLT ≅ IM > EM > VM. This indicates PFPOT provides best estimates while VM least estimates of speed of sound in present mixtures. The PFPOT is the only model which involves the interaction parameter. It is also observed that speeds of sound in IDPE + amines mixtures estimated using X_{12} obtained by the equation [12] in the PFPO theory are better than that when X_{12} = 0 was taken, while reverse is the case for oxolane + amines mixtures.

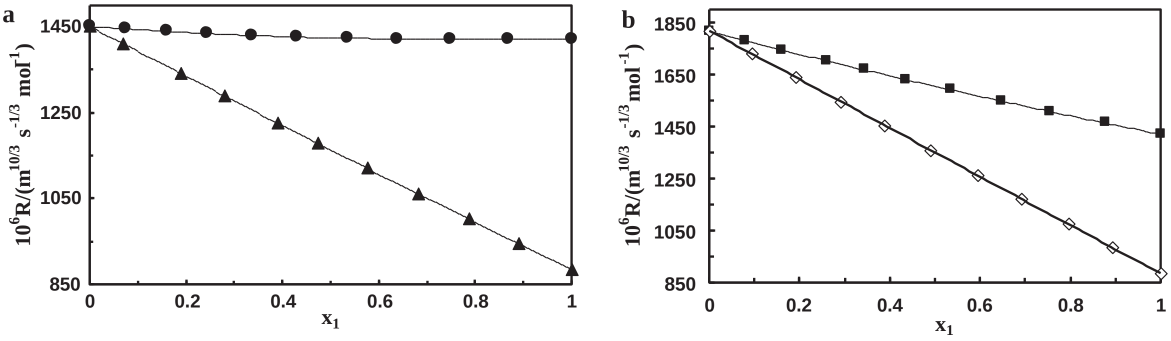

Fig. 5. Dependence of Rao's molar sound function on mole fraction of DIPE or oxolane at T = 303.15 K. (a) DIPE + DMA (●) and oxolane + DMA (▲). (b) DIPE + DEA (■) and oxolane + DEA (◊).

Table 10
Standard percentage deviation (σ (%)) between experimental and estimated speed of sound using empirical or theoretical relations for binary mixtures.

T /K	NM	VM	EM	IM	CFT	FLT	PFPOT	
							I[a]	II[b]
DIPE + DMA								
303.15	1.5	5.7	3.6	2.7	1.3	1.3	1.7 0.4	
313.15	2.1	6.5	4.3	2.5	1.1	1.3	1.5 0.2	
323.15	2.8	7.5	5.2	2.1	0.7	1.8	1.0	0.8
DIPE + DEA								
303.15	1.3	9.0	3.2	0.7	0.4	1.0	1.3	0.1
313.15	1.7	9.8	3.7	0.5	0.5	1.8	1.2	0.4
323.15	2.2	10.6	4.3	0.4	0.5	3.3	1.2	0.8
Oxolane + DMA								
303.15	0.1	5.7	0.3	0.9	0.7	1.2	0.2	0.4
313.15	0.5	6.5	0.8	1.4	1.1	0.9	0.6	0.9
323.15	0.9	7.0	1.2	1.8	0.8	1.5	0.9	1.2
Oxolane + DEA								
303.15	0.3	8.4	0.3	1.4	1.2	2.0	0.3	0.7
313.15	0.2	8.8	0.4	1.6	1.2	1.2	0.5	0.8
323.15	0.4	9.2	0.6	1.8	1.3	0.6	0.6	1.0
Aveg. σ (%)	1.2	7.9	2.3	1.5	0.9	1.5	0.9	0.6

[a] $X_{12} = 0$; X_{12} = from Eq. (12).

5. Conclusions

The magnitude of negative values of V_m^E, κ_S^E, and L_f^E and positive values of Z^E for binary mixtures of DIPE with DMA or DEA is much larger than that observed for oxolane mixtures at 303.15, 313.15, and 323.15 K. This difference is attributed to the effect due to the difference in free volume and internal pressure of involved components. The specific interactions between unlike molecules through dipole–dipole interactions exist and dominate over the dispersion interactions. The overall estimation ability of speed of sound in presently investigated mixtures follows the sequence: PFPOT > CFT > NM > FLT ≅ IM > EM > VM.

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