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Densities, Viscosities, Speeds of Sound, and Refractive Indices for Binary Mixtures of Diethylcarbonate, Acetophenone, and 1-Hexanol at (293.15, 303.15, 313.15, and 323.15) K for the Liquid Region and at Ambient Pressure

H. Iloukhani* and Z. Rostami

Faculty of Chemistry, University of Bu-Ali Sina, Hamadan, Iran

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Densities, viscosities, speeds of sound, and refractive indices of the binary systems formed by (diethylcarbonate + acetophenone), (diethylcarbonate + 1-hexanol), and (acetophenone + 1-hexanol) were measured at (293.15, 303.15, 313.15, and 323.15) K for the liquid region and at ambient pressure for the whole composition range. Deviations in viscosity ($\Delta\eta$) from the mole fraction average, deviations in isentropic compressibility ($\Delta\kappa_s$), and deviations in the refractive index (Δn_D) from the volume fraction average for the mixtures were derived from the experimental data. The binary data of $\Delta\eta$, $\Delta\kappa_s$, and Δn_D were correlated as a function of the mole fraction by using the Redlich−Kister-type function to determine the fitting parameters and the standard deviations. McAllister's four-body interaction model is used for correlating the kinematic viscosity of binary mixtures. The speeds of sound have been compared with values calculated from the free length theory (FLT) due to Jacobson and the collision factor theory due to Schaaffs. The experimental data of the constitute binaries are analyzed to discuss the nature and strength of intermolecular interactions in these mixtures.

Introduction

This paper is a continuation of our earlier work related to the study of thermodynamic and acoustic properties of binary mixtures.[1−5] In recent years, measurements of acoustic and transport properties have been adequately employed in understanding the nature of molecular systems and physicochemical behavior in liquid mixtures. The nonrectilinear behavior of the above-mentioned properties of liquid mixtures with changing mole or volume fractions is attributed to the difference in size of the molecules and strength of the interactions. Here, we have reported densities ρ, viscosities η, and refractive indices n_D for the binary systems formed by (diethylcarbonate + acetophenone), (diethylcarbonate + 1-hexanol), and (acetophenone + 1-hexanol) at (293.15, 303.15, 313.15, and 323.15) K for the liquid region and at ambient pressure for the whole composition range. The derived properties (deviations in the viscosity $\Delta\eta$, deviations in isentropic compressibility $\Delta\kappa_s$, and deviations in the refractive index Δn_D) in combination with other mixing properties provide valuable information for qualitatively analyzing the molecular interactions between molecules.

The deviation quantities of binary mixtures have been fitted to the Redlich−Kister[6] equation to determine the coefficients. The viscosity data for the binary mixtures were correlated to the semiempirical McAllister[7] model, and their parameters have been calculated. Theoretical models dealing with the liquid state, including the Jacobson free length theory (FLT)[8] and the Schaaff collision factor theory (CFT),[9] have been proposed for the prediction of the speeds of sound of the binary mixtures.

Experimental Section

Materials. The mole fraction purity of the molecules from Fluka were: diethylcarbonate (≥99.5 %), acetophenone (>99 %), and 1-hexanol (≥99 %). Compounds were purified by distillation using a 1 m fractionation column. The purity of each compound was ascertained by the constancy of the density and also from the refractive index, and their values were in good agreement with values found in the literature,[10−13] reported in Table 1. The purified compounds were stored in brown glass bottles and fractionally distilled immediately before use.

Table 1. Comparison of Measured Densities and Refractive Indices of the Pure Components with the Literature Values at 293.15 K

	$\rho/(\mathrm{g{\cdot}cm^{-3}})$		n_D	
component	this work	lit.	this work	lit
diethylcarbonate	0.97465	0.97468[10]	1.3845	1.3845[13]
acetophenone	1.02793	1.02810[11]	1.5336	1.5342[11]
1-hexanol	0.81884	0.81880[12]	1.4178	1.4181[11]

Apparatus and Procedure. The density of the compounds and their binary mixtures was measured with an Anton Paar DMA 4500 oscillating U-tube densitometer, operated in the static mode, and the uncertainties were estimated to be within $\pm 1 \times 10^{-5}$ g·cm^{-3}. The temperature in the cell was regulated to ±0.01 K with a solid-state thermostat. The apparatus was calibrated once a day with dry air and double-distilled freshly degassed water. Airtight stoppered bottles were used for the preparation of the mixtures. The mass of the dry bottle was first determined. The less volatile component of the mixture was introduced in the bottle, and the total mass was recorded. Subsequently, the other component was introduced, and the mass of the bottle along with the two components was determined. Each mixture was immediately used after it was well mixed by shaking. All the weightings were performed on an electronic balance (AB 204-N Mettler) accurate to 0.1 mg. The uncertainty in the mole fraction is estimated to be lower than $\pm 2 \times 10^{-4}$.

Dynamic viscosities were measured with an Ubbelohde viscometer. The equation for viscosity, according to Poiseuille's law, is

$$\eta = \rho\upsilon = \rho(kt - c/t) \qquad (1)$$

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* Corresponding author. E-mail: iloukhani@basu.ac.ir.

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Table 2. Experimental Densities ρ, Viscosities η, Speeds of Sound u, Refractive Indices n_D, Deviation in the Viscosity $\Delta\eta$, Deviation in the Isentropic Compressibility $\Delta\kappa_s$, and Deviation in the Refractive Index Δn_D, for x_1 Diethylcarbonate + (1 − x_1) Acetophenone at (293.15 to 323.15) K

	ρ	η	u		$\Delta\eta$	$\Delta\kappa_s$			ρ	η	u		$\Delta\eta$	$\Delta\kappa_s$	
x_1	g·cm^{-3}	mPa·s	m·s^{-1}	n_D	mPa·s	TPa^{-1}	Δn_D	x_1	g·cm^{-3}	mPa·s	m·s^{-1}	n_D	mPa·s	TPa^{-1}	Δn_D
T = 293.15 K								T = 313.15 K							
0.0511	1.02519	1.754	1443	1.5269	−0.016	18	0.0012	0.0511	1.00758	1.232	1365	1.5172	−0.010	24	0.0014
0.1014	1.02301	1.688	1405	1.5201	−0.031	31	0.0021	0.1014	1.00519	1.188	1326	1.5107	−0.021	41	0.0027
0.1501	1.02069	1.625	1377	1.5134	−0.044	39	0.0029	0.1501	1.00273	1.148	1292	1.5043	−0.029	57	0.0037
0.1996	1.01839	1.564	1347	1.5065	−0.055	49	0.0035	0.1996	1.00020	1.110	1263	1.4976	−0.034	71	0.0046
0.2465	1.01619	1.507	1322	1.5001	−0.064	58	0.0043	0.2465	0.99778	1.073	1238	1.4913	−0.041	83	0.0054
0.3041	1.01329	1.441	1297	1.4919	−0.071	66	0.0048	0.3041	0.99463	1.030	1212	1.4833	−0.046	95	0.0062
0.3467	1.01115	1.391	1280	1.4858	−0.078	71	0.0051	0.3467	0.99231	0.998	1196	1.4774	−0.050	102	0.0067
0.4001	1.00846	1.329	1265	1.4781	−0.085	73	0.0054	0.4001	0.98935	0.961	1180	1.4697	−0.052	107	0.0070
0.4507	1.00589	1.276	1251	1.4707	−0.086	74	0.0056	0.4507	0.98655	0.925	1168	1.4624	−0.055	108	0.0073
0.4888	1.00390	1.238	1242	1.4651	−0.086	74	0.0057	0.4888	0.98435	0.900	1160	1.4568	−0.055	108	0.0074
0.5506	1.00067	1.174	1231	1.4558	−0.087	71	0.0056	0.5506	0.98071	0.860	1153	1.4475	−0.055	101	0.0072
0.6038	0.99777	1.122	1222	1.4477	−0.084	69	0.0054	0.6038	0.97748	0.827	1152	1.4395	−0.053	88	0.0071
0.6498	0.99525	1.079	1218	1.4406	−0.080	62	0.0051	0.6498	0.97469	0.799	1151	1.4323	−0.051	78	0.0066
0.7104	0.99192	1.025	1215	1.4312	−0.073	52	0.0046	0.7104	0.97104	0.765	1150	1.4228	−0.045	63	0.0060
0.7513	0.98953	0.989	1213	1.4247	−0.068	45	0.0041	0.7513	0.96847	0.742	1149	1.4164	−0.042	54	0.0056
0.8078	0.98629	0.941	1210	1.4158	−0.057	35	0.0035	0.8078	0.96489	0.711	1148	1.4072	−0.035	42	0.0046
0.8522	0.98366	0.906	1208	1.4086	−0.048	28	0.0027	0.8522	0.96199	0.688	1147	1.3999	−0.029	32	0.0037
0.8993	0.98083	0.869	1207	1.4010	−0.037	19	0.0020	0.8993	0.95899	0.665	1146	1.3922	−0.021	22	0.0028
0.9488	0.97782	0.833	1206	1.3929	−0.022	9	0.0010	0.9488	0.95563	0.641	1145	1.3839	−0.012	11	0.0016
T = 303.15 K								T = 323.15 K							
0.0511	1.01640	1.455	1408	1.5221	−0.014	20	0.0013	0.0511	0.99879	1.057	1305	1.5123	−0.009	40	0.0015
0.1014	1.01409	1.402	1369	1.5154	−0.027	36	0.0023	0.1014	0.99627	1.023	1265	1.5060	−0.015	62	0.0030
0.1501	1.01177	1.352	1339	1.5089	−0.037	47	0.0033	0.1501	0.99368	0.991	1234	1.4998	−0.021	79	0.0043
0.1996	1.00935	1.302	1312	1.5022	−0.047	57	0.0042	0.1996	0.99108	0.959	1208	1.4932	−0.027	92	0.0053
0.2465	1.00704	1.257	1290	1.4958	−0.055	65	0.0049	0.2465	0.98856	0.929	1185	1.4869	−0.031	105	0.0061
0.3041	1.00402	1.203	1265	1.4877	−0.062	75	0.0055	0.3041	0.98524	0.893	1164	1.4790	−0.036	114	0.0069
0.3467	1.00178	1.165	1249	1.4817	−0.065	80	0.0059	0.3467	0.98280	0.868	1151	1.4731	−0.038	119	0.0075
0.4001	0.99899	1.118	1232	1.4740	−0.069	85	0.0063	0.4001	0.97974	0.837	1137	1.4656	−0.041	122	0.0080
0.4507	0.99627	1.075	1217	1.4666	−0.072	89	0.0064	0.4507	0.97677	0.809	1126	1.4583	−0.042	123	0.0083
0.4888	0.99418	1.042	1208	1.4610	−0.073	90	0.0065	0.4888	0.97449	0.787	1121	1.4527	−0.042	119	0.0083
0.5506	0.99073	0.993	1198	1.4518	−0.073	87	0.0065	0.5506	0.97076	0.755	1118	1.4435	−0.042	106	0.0083
0.6038	0.98773	0.952	1194	1.4437	−0.071	79	0.0063	0.6038	0.96747	0.727	1117	1.4353	−0.040	93	0.0079
0.6498	0.98506	0.918	1193	1.4366	−0.067	70	0.0060	0.6498	0.96454	0.704	1116	1.4282	−0.039	82	0.0076
0.7104	0.98154	0.875	1192	1.4271	−0.062	57	0.0054	0.7104	0.96063	0.676	1115	1.4186	−0.034	67	0.0068
0.7513	0.97905	0.848	1191	1.4206	−0.055	49	0.0048	0.7513	0.95786	0.656	1114	1.4121	−0.032	57	0.0063
0.8078	0.97568	0.811	1190	1.4116	−0.047	37	0.0041	0.8078	0.95413	0.631	1113	1.4029	−0.026	44	0.0053
0.8522	0.97291	0.784	1189	1.4043	−0.038	29	0.0032	0.8522	0.95107	0.612	1112	1.3957	−0.022	33	0.0045
0.8993	0.96991	0.755	1188	1.3966	−0.029	20	0.0023	0.8993	0.94776	0.592	1111	1.3878	−0.016	23	0.0033
0.9488	0.96678	0.727	1187	1.3884	−0.017	10	0.0013	0.9488	0.94432	0.573	1110	1.3791	−0.009	11	0.0017

where k and c are the viscometer constants and t, η, and υ are the efflux time, dynamic viscosity, and kinematic viscosity, respectively. The dynamic viscosity was reproducible to within $\pm 2 \times 10^{-3}$ mPa·s. The viscometer was suspended in a thermostated water bath maintained to ±0.01 K.

The speeds of sound in the pure compounds and their binary mixtures were measured with a multifrequency ultrasonic interferometer supplied by Mittal Enterprise, New Delhi, with an accuracy of ±1 m·s^{-1}. In this work, 3 MHz frequency was employed.

Refractive indices were measured using a digital Abbe-type refractometer. The uncertainty of refractive index measurement was estimated to be less than ±0.0001 units.

Results and Discussion

Dynamic and Kinematic Viscosities. The measured ρ and η values for binary systems at (293.15, 303.15, 313.15, and 323.15) K are listed in Tables 2 to 4.

The viscosity deviations $\Delta\eta$[14] for binary mixtures can be calculated as

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

where η and η_i are the viscosities of the binary mixture and pure components. The computed values of $\Delta\eta$ for binary mixtures are also recorded in Tables 2 to 4 and are graphically represented in Figures 1a to 3a. The deviations in viscosity $\Delta\eta$ for the three systems of (diethylcarbonate + acetophenone), (diethylcarbonate + 1-hexanol), and (acetophenone + 1-hexanol) are negative over the entire range of composition and become less negative with increasing temperature from (293.15 to 323.15) K. This reveals that the strength of specific interaction is not the only factor influencing the viscosity deviation of liquid mixtures. The molecular size and shape of the components also play an equally important role.

Speeds of Sound and Deviations in Isentropic Compressibility. Isentropic compressibility κ_s was calculated using the Newton−Laplace equation

$$\kappa_s = 1/\rho u^2 \tag{3}$$

Deviations in isentropic compressibilities $\Delta\kappa_s$ of the n-component were calculated using the following relation

$$\Delta\kappa_s = \kappa_s - \sum_i (\kappa_{s,i}^*)\phi_i \tag{4}$$

Table 3. Experimental Densities ρ, Viscosities η, Speeds of Sound u, Refractive Indices n_D, Deviation in the Viscosity $\Delta\eta$, Deviation in the Isentropic Compressibility $\Delta\kappa_s$, and Deviation in the Refractive Index Δn_D, for x_1 Diethylcarbonate + $(1 - x_1)$ 1-Hexanol at (293.15 to 323.15) K

x_1	ρ g·cm^{-3}	η mPa·s	u m·s^{-1}	n_D	$\Delta\eta$ mPa·s	$\Delta\kappa_s$ TPa^{-1}	Δn_D
			$T = 293.15$ K				
0.0502	0.82596	4.731	1301	1.4168	−0.333	1373	0.0006
0.1123	0.83486	4.164	1291	1.4152	−0.621	7	0.0010
0.1499	0.84029	3.715	1285	1.4143	−0.902	10	0.0014
0.2103	0.84910	3.178	1275	1.4125	−1.168	14	0.0015
0.2508	0.85505	2.837	1268	1.4111	−1.327	17	0.0015
0.3121	0.86419	2.382	1259	1.4089	−1.508	20	0.0013
0.3576	0.87102	2.078	1252	1.4071	−1.607	22	0.0010
0.4113	0.87915	1.757	1245	1.4049	−1.687	24	0.0006
0.4632	0.88708	1.488	1239	1.4028	−1.724	24	0.0002
0.5111	0.89451	1.247	1234	1.4008	−1.749	24	−0.0002
0.5520	0.90088	1.115	1230	1.3991	−1.698	24	−0.0006
0.6008	0.90859	0.956	1226	1.3972	−1.638	23	−0.0008
0.6498	0.91639	0.863	1222	1.3953	−1.511	22	−0.0011
0.7108	0.92619	0.859	1219	1.3929	−1.242	18	−0.0014
0.7554	0.93345	0.840	1216	1.3912	−1.060	16	−0.0016
0.8112	0.94265	0.831	1213	1.3894	−0.819	12	−0.0015
0.8501	0.94913	0.825	1211	1.3883	−0.651	10	−0.0013
0.9002	0.95750	0.818	1208	1.3869	−0.433	8	−0.0010
0.9555	0.96689	0.803	1206	1.3855	−0.200	3	−0.0005
			$T = 303.15$ K				
0.0502	0.81855	3.486	1267	1.4131	−0.228	8	0.0008
0.1123	0.82716	3.030	1256	1.4115	−0.486	14	0.0013
0.1499	0.83248	2.796	1249	1.4106	−0.602	18	0.0016
0.2103	0.84104	2.403	1240	1.4088	−0.803	22	0.0018
0.2508	0.84679	2.165	1234	1.4074	−0.913	25	0.0018
0.3121	0.85569	1.859	1225	1.4051	−1.024	29	0.0016
0.3576	0.86223	1.621	1217	1.4033	−1.118	34	0.0013
0.4113	0.87021	1.414	1210	1.4011	−1.155	37	0.0009
0.4632	0.87788	1.220	1204	1.3989	−1.184	39	0.0005
0.5111	0.88513	1.063	1200	1.3969	−1.189	39	0.0002
0.5520	0.89135	0.951	1197	1.3952	−1.172	38	−0.0001
0.6008	0.89887	0.844	1194	1.3931	−1.125	36	−0.0006
0.6498	0.90642	0.811	1193	1.3911	−1.002	31	−0.0009
0.7108	0.91593	0.804	1192	1.3887	−0.816	26	−0.0012
0.7554	0.92308	0.774	1191	1.3870	−0.704	22	−0.0013
0.8112	0.93203	0.748	1190	1.3851	−0.554	17	−0.0013
0.8501	0.93829	0.730	1189	1.3838	−0.447	13	−0.0012
0.9002	0.94657	0.724	1188	1.3823	−0.295	9	−0.0010
0.9555	0.95577	0.713	1187	1.3809	−0.131	4	−0.0005
			$T = 313.15$ K				
0.0502	0.81104	2.621	1252	1.4089	−0.162	13	0.0009
0.1123	0.81937	2.339	1236	1.4075	−0.303	24	0.0016
0.1499	0.82454	2.115	1224	1.4065	−0.442	33	0.0019
0.2103	0.83278	1.856	1211	1.4047	−0.563	41	0.0022
0.2508	0.83835	1.687	1203	1.4034	−0.639	45	0.0023
0.3121	0.84697	1.466	1192	1.4011	−0.721	50	0.0021
0.3576	0.85342	1.319	1185	1.3992	−0.765	52	0.0017
0.4113	0.86112	1.160	1177	1.3969	−0.801	54	0.0013
0.4632	0.86859	1.031	1170	1.3947	−0.811	55	0.0009
0.5111	0.87561	0.912	1165	1.3927	−0.821	54	0.0005
0.5520	0.88155	0.826	1162	1.3909	−0.814	51	0.0002
0.6008	0.88891	0.744	1158	1.3888	−0.786	49	−0.0002
0.6498	0.89635	0.689	1155	1.3867	−0.729	45	−0.0006
0.7108	0.90565	0.610	1153	1.3842	−0.669	37	−0.0010
0.7554	0.91255	0.607	1152	1.3825	−0.570	31	−0.0011
0.8112	0.92116	0.602	1151	1.3805	−0.448	23	−0.0012
0.8501	0.92749	0.606	1150	1.3792	−0.356	17	−0.0011
0.9002	0.93557	0.610	1148	1.3776	−0.238	12	−0.0009
0.9555	0.94449	0.615	1146	1.3762	−0.106	5	−0.0004
			$T = 323.15$ K				
0.0502	0.80329	2.060	1204	1.4049	−0.126	15	0.0010
0.1123	0.81130	1.825	1184	1.4036	−0.254	34	0.0019
0.1499	0.81620	1.689	1172	1.4026	−0.326	46	0.0022
0.2103	0.82420	1.488	1160	1.4007	−0.423	54	0.0024
0.2508	0.82957	1.367	1153	1.3994	−0.474	58	0.0026
0.3121	0.83788	1.220	1143	1.3971	−0.516	64	0.0024
0.3576	0.84405	1.093	1137	1.3953	−0.565	66	0.0022
0.4113	0.85148	0.970	1131	1.3931	−0.595	66	0.0019
0.4632	0.85873	0.857	1126	1.3909	−0.619	66	0.0016
0.5111	0.86559	0.765	1123	1.3887	−0.629	62	0.0011
0.5520	0.87151	0.689	1121	1.3868	−0.635	58	0.0006
0.6008	0.87865	0.607	1119	1.3846	−0.633	53	0.0002
0.6498	0.88585	0.568	1117	1.3825	−0.588	48	−0.0002
0.7108	0.89496	0.532	1115	1.3798	−0.519	41	−0.0007
0.7554	0.90177	0.535	1114	1.3779	−0.439	34	−0.0010
0.8112	0.91041	0.537	1113	1.3758	−0.341	26	−0.0011
0.8501	0.91642	0.540	1112	1.3745	−0.271	21	−0.0010
0.9002	0.92430	0.544	1111	1.3729	−0.181	14	−0.0008
0.9555	0.93324	0.548	1110	1.3714	−0.082	6	−0.0003

(where $\kappa^*_{s,i}$, κ_s, and ϕ_i are the isentropic compressibilities of pure components, mixtures, and volume fraction average, respectively) and

$$\phi_i = \frac{x_i V_i}{\sum_{i=1}^{N} x_i V_i} \tag{5}$$

(where V and V_i are the molar volume of the mixture and the molar volume of pure component i, respectively).

The corresponding values for isentropic compressibility, κ_s, and deviation in isentropic compressibilities, $\Delta\kappa_s$, of the binary mixtures measured at (293.15, 303.15, 313.15, and 323.15) K are also presented in Tables 2 to 4. The deviations in isentropic compressibility were plotted against mole fraction average and showed in Figures 1b to 3b.

The values of $\Delta\kappa_s$ are positive for the systems (diethylcarbonate + acetophenone) and (diethylcarbonate + 1-hexanol), become more positive with increasing temperature from (293.15 to 323.15) K, are negative for the system (acetophenone + 1-hexanol), and become more negative with increasing temperature over the whole composition range. The positive $\Delta\kappa_s$ values of these binary mixtures are due to the dispersion interactions between unlike molecules, and the negative $\Delta\kappa_s$ values of the (acetophenone + 1-hexanol) binary mixture are due to the polar interactions between the hydrogen atom of the −OH group of 1-hexanol and oxygen atoms of acetophenone.

Deviations in Refractive Index. The deviation in the refractive index, Δn_D, was calculated from the volume fraction average as suggested by Brocos et al.[15] and is given by

$$\Delta n_D = n_D - \sum_{i=1}^{N} \phi_i n_{D_i} \tag{6}$$

where $n_{D,}$ n_{Di}, and ϕ_i are the refractive index of the mixture, the refractive index of pure component i, and the volume fraction of pure component i, respectively. The corresponding values for deviations in refractive index, Δn_D, of the binary mixtures measured at (293.15, 303.15, 313.15, and 323.15) K are also presented in Tables 2 to 4. The deviations in refractive index were plotted against mole fraction average and are shown in Figures 1c to 3c.

For the whole composition range, the Δn_D values are positive for systems formed by (diethylcarbonate + acetophenone) and (acetophenone + 1-hexanol) and become more positive with increasing temperature from (293.15 to 323.15) K. The deviations in refractive index of (diethylcarbonate + 1-hexanol) show an S-shaped dependence on composition with positive values

Table 4. Experimental Densities ρ, Viscosities η, Speeds of Sound u, Refractive Indices n_D, Deviation in the Viscosity $\Delta\eta$, Deviation in the Isentropic Compressibility $\Delta\kappa_s$, and Deviation in the Refractive Index Δn_D, for x_1 Acetophenone + (1 − x_1) 1-Hexanol at (293.15 to 323.15) K

x_1	ρ g·cm^{-3}	η mPa·s	u m·s^{-1}	n_D	$\Delta\eta$ mPa·s	$\Delta\kappa_s$ TPa^{-1}	Δn_D	x_1	ρ g·cm^{-3}	η mPa·s	u m·s^{-1}	n_D	$\Delta\eta$ mPa·s	$\Delta\kappa_s$ TPa^{-1}	Δn_D
			T = 293.15 K								T = 313.15 K				
0.0498	0.82869	4.831	1.4234	1316	−0.286	−2	0.0002	0.0498	0.81397	2.684	1277	1.4153	−0.133	−5	0.0003
0.1009	0.83884	4.397	1.4291	1323	−0.543	−4	0.0003	0.1009	0.82385	2.481	1285	1.4210	−0.253	−10	0.0005
0.1517	0.84899	3.997	1.4348	1331	−0.767	−7	0.0004	0.1517	0.83375	2.292	1293	1.4267	−0.359	−14	0.0006
0.2106	0.86083	3.605	1.4415	1341	−0.954	−11	0.0005	0.2106	0.84532	2.104	1303	1.4334	−0.452	−19	0.0009
0.2474	0.86827	3.310	1.4457	1348	−1.121	−13	0.0006	0.2474	0.85258	1.977	1310	1.4376	−0.519	−23	0.0011
0.3028	0.87953	2.986	1.4521	1358	−1.254	−15	0.0008	0.3028	0.86361	1.812	1320	1.4439	−0.594	−26	0.0012
0.3426	0.88767	2.744	1.4567	1365	−1.358	−17	0.0009	0.3426	0.87157	1.697	1328	1.4484	−0.644	−29	0.0013
0.3978	0.89901	2.491	1.4631	1375	−1.420	−18	0.0010	0.3978	0.88270	1.580	1339	1.4547	−0.672	−33	0.0014
0.4512	0.91005	2.266	1.4693	1385	−1.459	−19	0.0011	0.4512	0.89355	1.483	1350	1.4608	−0.682	−36	0.0015
0.4985	0.91988	2.085	1.4748	1394	−1.477	−19	0.0012	0.4985	0.90323	1.396	1360	1.4662	−0.693	−38	0.0015
0.5526	0.93120	1.930	1.4811	1404	−1.444	−19	0.0012	0.5526	0.91438	1.322	1370	1.4724	−0.679	−39	0.0016
0.6071	0.94267	1.798	1.4874	1415	−1.387	−19	0.0011	0.6071	0.92570	1.263	1380	1.4786	−0.649	−39	0.0015
0.6491	0.95155	1.744	1.4923	1424	−1.295	−19	0.0011	0.6491	0.93449	1.255	1387	1.4834	−0.589	−38	0.0014
0.7101	0.96453	1.650	1.4994	1437	−1.177	−18	0.0009	0.7101	0.94735	1.243	1398	1.4903	−0.503	−37	0.0012
0.7485	0.97275	1.664	1.5039	1445	−1.030	−16	0.0009	0.7485	0.95550	1.252	1404	1.4947	−0.431	−35	0.0011
0.8019	0.98423	1.673	1.5101	1457	−0.837	−15	0.0007	0.8019	0.96690	1.264	1408	1.5008	−0.332	−29	0.0009
0.8521	0.99509	1.691	1.516	1467	−0.644	−12	0.0005	0.8521	0.97772	1.266	1409	1.5066	−0.249	−20	0.0007
0.9045	1.00650	1.710	1.5222	1476	−0.444	−8	0.0004	0.9045	0.98909	1.271	1411	1.5126	−0.159	−12	0.0005
0.9554	1.01763	1.727	1.5283	1485	−0.250	−4	0.0002	0.9554	1.00023	1.276	1416	1.5185	−0.071	−6	0.0002
			T = 303.15 K								T = 323.15 K				
0.0498	0.82141	3.559	1.4196	1285	−0.196	−3	0.0002	0.0498	0.80647	2.119	1230	1.4113	−0.094	−9	0.0004
0.1009	0.83148	3.262	1.4253	1293	−0.373	−7	0.0004	0.1009	0.81628	1.970	1238	1.4170	−0.183	−14	0.0007
0.1517	0.84154	2.989	1.4310	1302	−0.526	−11	0.0005	0.1517	0.82611	1.831	1247	1.4227	−0.263	−20	0.0009
0.2106	0.85328	2.725	1.4377	1313	−0.651	−16	0.0008	0.2106	0.83759	1.690	1259	1.4293	−0.334	−27	0.0011
0.2474	0.86066	2.511	1.4419	1320	−0.777	−18	0.0009	0.2474	0.84480	1.585	1266	1.4335	−0.395	−31	0.0013
0.3028	0.87183	2.306	1.4482	1332	−0.852	−23	0.0010	0.3028	0.85574	1.480	1278	1.4398	−0.436	−37	0.0015
0.3426	0.87989	2.148	1.4528	1340	−0.915	−25	0.0012	0.3426	0.86365	1.395	1287	1.4443	−0.474	−41	0.0016
0.3978	0.89114	1.961	1.4591	1351	−0.972	−27	0.0013	0.3978	0.87471	1.307	1298	1.4506	−0.497	−45	0.0018
0.4512	0.90209	1.805	1.4653	1362	−1.002	−29	0.0014	0.4512	0.88548	1.236	1308	1.4567	−0.504	−47	0.0019
0.4985	0.91185	1.692	1.4707	1371	−1.003	−30	0.0014	0.4985	0.89510	1.169	1316	1.4621	−0.516	−47	0.0020
0.5526	0.92308	1.583	1.4769	1382	−0.984	−30	0.0014	0.5526	0.90619	1.118	1326	1.4682	−0.503	−47	0.0020
0.6071	0.93447	1.488	1.4832	1393	−0.951	−29	0.0014	0.6071	0.91744	1.075	1335	1.4743	−0.482	−46	0.0018
0.6491	0.94329	1.442	1.4880	1402	−0.897	−29	0.0013	0.6491	0.92617	1.069	1342	1.4790	−0.438	−45	0.0017
0.7101	0.95619	1.431	1.4950	1414	−0.764	−27	0.0011	0.7101	0.93895	1.073	1351	1.4858	−0.362	−41	0.0015
0.7485	0.96435	1.426	1.4994	1421	−0.679	−25	0.0010	0.7485	0.94706	1.075	1356	1.4901	−0.315	−38	0.0013
0.8019	0.97577	1.455	1.5056	1430	−0.523	−21	0.0008	0.8019	0.95840	1.073	1361	1.4962	−0.254	−32	0.0011
0.8521	0.98656	1.465	1.5114	1439	−0.395	−17	0.0006	0.8521	0.96916	1.067	1365	1.5019	−0.201	−25	0.0009
0.9045	0.99788	1.480	1.5175	1445	−0.255	−11	0.0004	0.9045	0.98048	1.076	1370	1.5079	−0.130	−18	0.0007
0.9554	1.00895	1.496	1.5235	1451	−0.120	−4	0.0002	0.9554	0.99156	1.086	1372	1.5137	−0.060	−9	0.0004

in the 1-hexanol-rich region and negative values at the opposite extreme.

Each set of results was fitted using a Redlich−Kister[6] polynomial which for binary mixtures is

$$Y = x_1(1 - x_1)\sum_{k=0}^{N} A_k(2x_1 - 1)^k \tag{7}$$

where Y = ($\Delta\eta$ or $\Delta\kappa_s$ or Δn_D) and x_1 is the mole fraction of the first component. The values of coefficients A_k were determined by a multiple regression analysis based on the least-squares method.

In each case, the optimum number of coefficients was ascertained from an examination of the variation of standard deviation σ with

$$\sigma = [\sum (Y_{\text{exptl}} - Y_{\text{calcd}})^2/(n - p)]^{1/2} \tag{8}$$

where Y_{calcd} is the calculated values of the property Y and n and p are the number of experimental points and the number of parameters retained in the respective equations, respectively. A_k and values of $\Delta\eta$, $\Delta\kappa_s$, or Δn_D for binary mixtures are represented in Table 5.

Theoretical. McAllister's four-body interaction model[7] is widely used for correlating the kinematic viscosity of binary mixtures with mole fraction. The four-body model is defined as

$$\begin{aligned}\ln \nu = {} & x_1^4 \ln \nu_1 + 4x_1^3x_2 \ln \nu_{1112} + 6x_1^2x_2^2 \ln \nu_{1122} + \\ & 4x_1x_2^3 \ln \nu_{2221} + x_2^4 \ln \nu_2 - \ln\left(x_1 + \frac{x_2M_2}{M_1}\right) + \\ & 4x_1^3x_2 \ln\left(\frac{3 + M_2/M_1}{4}\right) + 6x_1^2x_2^2 \ln\left(\frac{1 + M_2/M_1}{2}\right) \\ & + 4x_1x_2^3 \ln\left(\frac{1 + 3M_2/M_1}{4}\right) + x_2^4 \ln\left(\frac{M_2}{M_1}\right)\end{aligned} \tag{9}$$

where υ, υ_1, and υ_2 are the kinematic viscosities of the mixture and the viscosities of pure components 1 and 2, respectively. υ_{12}, υ_{21}, υ_{1112}, υ_{1122}, and υ_{2221} are the model parameters. Table 6 records the calculated results with the standard deviations defined as eq 9.

The empirical and semiempirical theories of Jacobson and Schaaff were used to obtain speeds of sound in the binary systems.

(i) Jacobson Free Length Theory (FLT).[8]

$$u = \frac{K}{L\rho^{1/2}} \tag{10}$$

Here, K is the temperature-dependent constant whose value at

Table 5. Parameters of the Redlich−Kister Equation and the Standard Deviations for Binary Mixtures at (293.15 to 323.15) K

system	A_0	A_1	A_2	A_3	A_4	σ
Diethylcarbonate + Acetophenone						
$T = 293.15$ K						
$\Delta\eta$/(mPa·s)	−0.3435	0.0067	−0.0493	−0.0849		1.3×10^{-3}
$\Delta\kappa_s$/(TPa^{-1})	298.58	−46.331	−121.61	−61.515	121.34	1.2
Δn_D	0.0225	0.0006	−0.0001	−0.003		6.7×10^{-5}
$T = 303.15$ K						
$\Delta\eta$/(mPa·s)	−0.2897	0.0059	−0.029	−0.0416		9.8×10^{-4}
$\Delta\kappa_s$/(TPa^{-1})	352.57	−74.86	−223.26	−51.306	219.31	1.1
Δn_D	0.0261	−0.00002	−0.00005	−0.00005		5.6×10^{-5}
$T = 313.15$ K						
$\Delta\eta$/(mPa·s)	−0.2196	0.0034	−0.0118	−0.0232		7.4×10^{-4}
$\Delta\kappa\ s$/(TPa^{-1})	421.62	−160.89	−269.46	28.956	241.67	1.3
Δn_D	0.0295	−0.0007	−0.0025	0.0031	0.0053	6.5×10^{-5}
$T = 323.15$ K						
$\Delta\eta$/(mPa·s)	−0.1706	−0.0015	0.0283	−0.0023	−0.0546	5.1×10^{-4}
$\Delta\kappa_s$/(TPa^{-1})	463.66	−222.08	−229.58	−108.04	378.18	2.5
Δn_D	0.0333	−0.00006	0.0009	0.0031		8.3×10^{-5}
Diethylcarbonate + 1-Hexanol						
$T = 293.15$ K						
$\Delta\eta$/(mPa·s)	−7.0176	1.0654	3.0328	0.1924	−1.8135	2.9×10^{-2}
$\Delta\kappa_s$/(TPa^{-1})	98.627	0.3755	−56.615	−1.721	37.812	0.5
Δn_D	−0.0003	−0.017	0.0007	0.0046		6.3×10^{-5}
$T = 303.15$ K						
$\Delta\eta$/(mPa·s)	−4.7694	0.8605	1.8876	0.135	−1.1158	1.8×10^{-2}
$\Delta\kappa_s$/(TPa^{-1})	154.79	−1.9497	−142.64	−46.325	141.5	0.6
Δn_D	0.0009	−0.0165	0.0015	0.0011		4.9×10^{-5}
$T = 313.15$ K						
$\Delta\eta$/(mPa·s)	−3.3142	0.174	0.3689	0.3523	0.186	1.2×10^{-2}
$\Delta\kappa_s$/(TPa^{-1})	217.14	−63.29	−68.103	−26.253	48.529	0.8
Δn_D	0.0024	−0.0186	0.0022	0.0034		6.4×10^{-5}
$T = 323.15$ K						
$\Delta\eta$/(mPa·s)	−2.5472	−0.1022	0.4935	0.6733	−0.2029	1.2×10^{-2}
$\Delta\kappa_s$/(TPa^{-1})	253.16	−146.58	−14.344	52.847		1.3
Δn_D	0.0048	−0.0197	−0.005	0.0043	0.0093	6.8×10^{-5}
Acetophenone + 1-Hexanol						
$T = 293.15$ K						
$\Delta\eta$/(mPa·s)	−5.9653	0.6091	1.8343	−0.4025	−2.1108	2.2×10^{-2}
$\Delta\kappa_s$/(TPa^{-1})	−77.38	−15.449	−10.355	−19.744	29.864	0.432
Δn_D	0.0047	0.0016	−0.0048	−0.0015	0.0054	3.3×10^{-5}
$T = 303.15$ K						
$\Delta\eta$/(mPa·s)	−4.0613	0.3435	1.2025	0.5292	−0.6029	1.3×10^{-2}
$\Delta\kappa_s$/(TPa^{-1})	−117.68	−37.325	−22.143	16.732	81.382	0.672
Δn_D	0.0057	0.0007	−0.0036	−0.0006	0.0025	3.2×10^{-5}
$T = 313.15$ K						
$\Delta\eta$/(mPa·s)	−2.792	0.4002	1.1498	0.2955	−0.6009	8.1×10^{-3}
$\Delta\kappa_s$/(TPa^{-1})	−152.1	−76.376	7.289	74.851	39.417	0.911
Δn_D	0.0062	0.001	−0.0027	−0.0018	0.0024	4.6×10^{-5}
$T = 323.15$ K						
$\Delta\eta$/(mPa·s)	−2.0577	0.3531	0.7433	−0.0433	−0.3962	8.3×10^{-3}
$\Delta\kappa_s$/(TPa^{-1})	−193.05	−39.096	69.104	24.924	−90.411	0.996
Δn_D	0.0079	0.0001	−0.006	0.0003	0.009	3.2×10^{-5}

(293.15, 303.15, 313.15, and 323.15) K is 618, 631, 642, and 652, respectively. ρ is the density of the mixture. L_f is the intermolecular free length of the mixture. Here

$$L_f = \frac{2\left(1/\rho - \sum_{i=1}^{2} w_i V_{0i}/M_i\right)}{\sum_{i=1}^{2} w_i Y_i/M_i} \tag{11a}$$

where M_i is the molecular weight of component i; w_i is the weight fraction of component i; and Y_i and V_{0i} are the molar surface factor area per mole and the molar volumes at a temperature of absolute zero of component i, respectively. Y_i and V_{0i} are given by

$$Y_i = (36\pi N_A V_{0i}^{\ 2})^{1/3} \tag{11b}$$

and

$$V_{0i} = V^0\left(1 - \frac{T}{T_c}\right)^{0.3} \tag{11c}$$

Table 6. Parameters of McAllister's Four-Body Interaction Equation and Standard Deviations for Kinematic Viscosities at (293.15 to 323.15) K

system	T/K	υ_{1112}/(mm^2·s^{-1})	υ_{1122}/(mm^2·s^{-1})	υ_{2221}/(mm^2·s^{-1})	σ/(mm^2·s^{-1})
diethylcarbonate + acetophenone	293.15	0.98758	1.24415	1.48402	1.8×10^{-3}
	303.15	0.85584	1.05071	1.24774	8.0×10^{-4}
	313.15	0.75927	0.91415	1.07540	8.2×10^{-4}
	323.15	0.68280	0.80441	0.94057	3.5×10^{-4}
diethylcarbonate + 1-hexanol	293.15	0.82613	0.81111	4.87718	0.1106
	303.15	0.82777	0.72603	3.48943	0.0703
	313.15	0.52298	0.87490	2.30416	0.0276
	323.15	0.48369	0.69059	1.96328	0.0377
acetophenone + 1-hexanol	293.15	1.49470	1.72795	4.28630	0.0418
	303.15	1.44818	1.30316	3.30342	0.0360
	313.15	1.36539	1.07572	2.57979	0.0241
	323.15	1.15364	0.96898	2.04333	0.0154

Table 7. Molar Volume (V^0), Molar Volume at Absolute Zero (V_0), Available Volume ($V_a = V^0 - V_0$), Intermolecular Free Length (L_f), Molar Surface Factor (Y), Molecular Radius (r), Molar Actual Volume of Molecule (B), and Collision Factor (S) of the Pure Components at (293.15 to 323.15) K

compound	T	V^0	V_0	V_a	B	Y	r	L_f	S
	K	10^{-6} m^3·mol^{-1}				m^2·mol^{-1}	10^{-10} m		
diethylcarbonate	293.15	121.202	103.902	17.300	54.530	66.549	2.7865	0.5199	1.6726
	303.15	122.617	104.387	18.230	54.593	67.262	2.7876	0.5420	1.6649
	313.15	124.076	104.881	19.195	54.641	66.749	2.7884	0.5751	1.6236
	323.15	125.585	105.386	20.199	54.671	66.642	2.7889	0.6062	1.5922
acetophenone	293.15	116.946	99.665	17.281	59.496	84.688	2.8686	0.4081	1.8354
	303.15	117.942	99.7838	18.158	59.574	84.754	2.8699	0.4285	1.8053
	313.15	118.956	99.8919	19.064	59.641	84.694	2.8710	0.4502	1.7689
	323.15	119.988	99.9894	19.999	59.697	84.284	2.8719	0.4746	1.7248
1-hexanol	293.15	124.786	102.540	22.246	59.444	85.344	2.8678	0.5213	1.7187
	303.15	125.889	102.456	23.432	59.518	85.513	2.8690	0.5480	1.6895
	313.15	127.026	102.361	24.665	59.589	87.454	2.8701	0.5641	1.6907
	323.15	128.207	102.257	25.950	59.659	86.697	2.8712	0.5986	1.6386

Table 8. Standard Deviations of Predicted Speeds of Sound and Isentropic Compressibilities, by Means of CFT and FLT of Binary Mixtures at (293.15 to 323.15) K

	T/K	CFT	FLT
	Diethylcarbonate + Acetophenone		
σ/(m·s^{-1})	293.15	82	103
	303.15	85	106
	313.15	91	112
	323.15	94	114
	Diethylcarbonate + 1-Hexanol		
σ/(m·s^{-1})	293.15	12	3
	303.15	18	3
	313.15	26	11
	323.15	27	11
	Acetophenone + 1-Hexanol		
σ/(m·s^{-1})	293.15	4	4
	303.15	5	12
	313.15	15	25
	323.15	17	28

where V^0 and T_c are, respectively, the molar volume of a pure component at temperature T and the critical temperature.

(ii) Schaaff Collision Factor Theory (CFT).[9]

$$u = u_\infty \frac{\left(\sum_{i=1}^{n} x_i S_i\right)\left(\sum_{i=1}^{n} x_i B_i\right)}{V} = u_\infty S r_f \qquad (12)$$

where $u_\infty = 1600$ m·s^{-1}, $S =$ the collision factor, and $r_f = B/V$, the space filling factor. B is the molar actual volume of the molecule.

B_i can be evaluated as

$$B_i = \frac{4}{3}\pi r^3 N_A \qquad (13a)$$

where r is the molecular radius of the pure component. The molecular radius is calculated as

$$r = \left[\frac{3b}{16\pi N_A}\right]^{1/3} \qquad (13b)$$

where b is the Van der Waals constant.

Values of parameters for pure components employed in and evaluated from the FLT and CFT are listed in Table 7. The theoretical speeds of sound, u, for the binary mixtures over the entire range of compositions were estimated. The values of the standard deviations are included in Table 8. The study of standard deviations in Table 8 reveals that the CFT model is suitable for (acetophenone + 1-hexanol) and that the FLT model gives the better results for (diethylcarbonate + 1-hexanol). For (diethylcarbonate + acetophenone), neither the CFT model nor the FLT model is matched with the experimental results.

Conclusions

Densities, viscosities, and refractive indices for binary mixtures consisting of (diethylcarbonate + acetophenone), (diethylcarbonate + 1-hexanol), and (acetophenone + 1-hexanol) have been measured at (293.15, 303.15, 313.15, and 323.15) K, and the corresponding deviations in the viscosity $\Delta\eta$, deviations in isentropic compressibility $\Delta\kappa_S$, and deviations in the refractive index Δn_D have been calculated to provide valuable information for qualitatively analyzing the molecular interactions between molecules.

The negative viscosity deviations suggest that in these mixtures the forces between pairs of unlike molecules are less

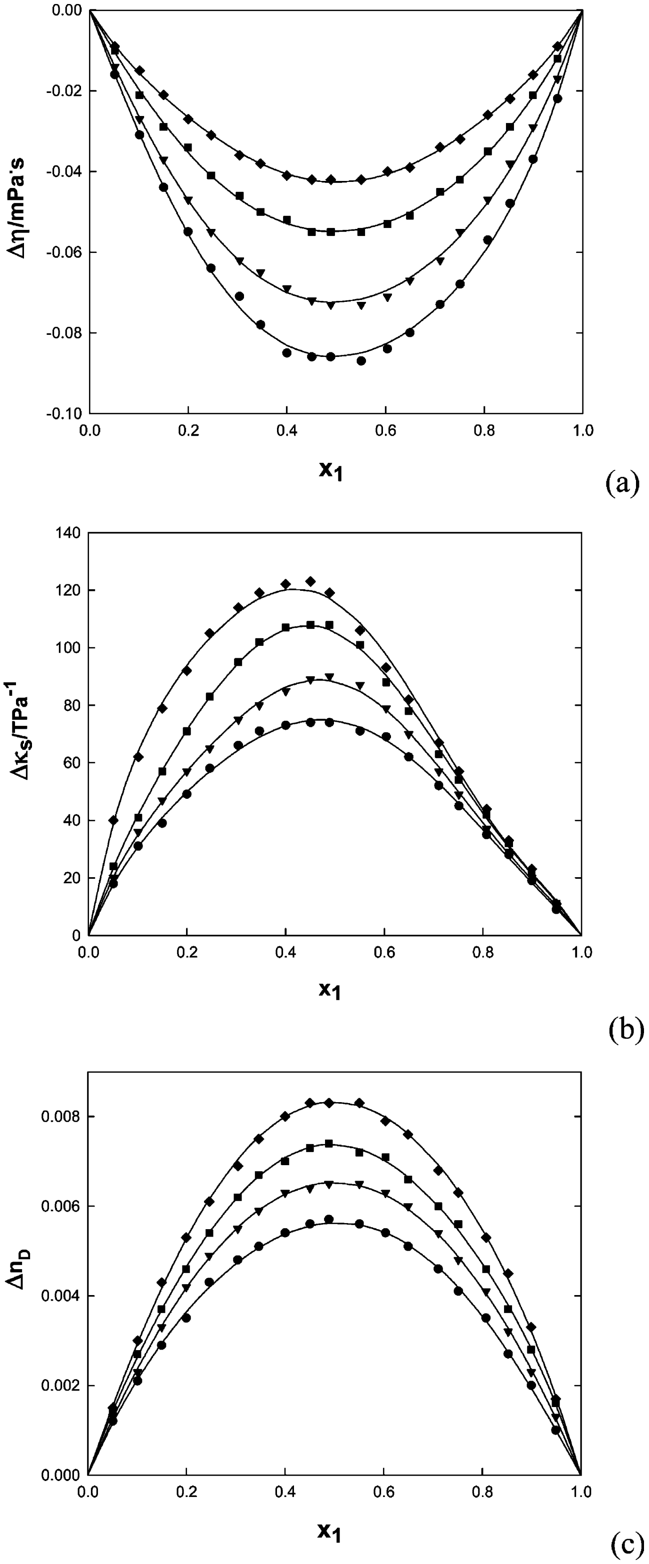

Figure 1. (a) Variation of the deviation in the viscosity $\Delta\eta$. (b) Variation of the deviation in the isentropic compressibility $\Delta\kappa_S$. (c) Variation of the deviation in the refractive index Δn_D, with mole fraction x_1 for x_1 diethylcarbonate + $(1 - x_1)$ acetophenone at the temperatures: ●, 293.15 K; ▼, 303.15 K; ■, 313.15 K; ◆, 323.15 K. Solid curves were calculated from the Redlich−Kister equation.

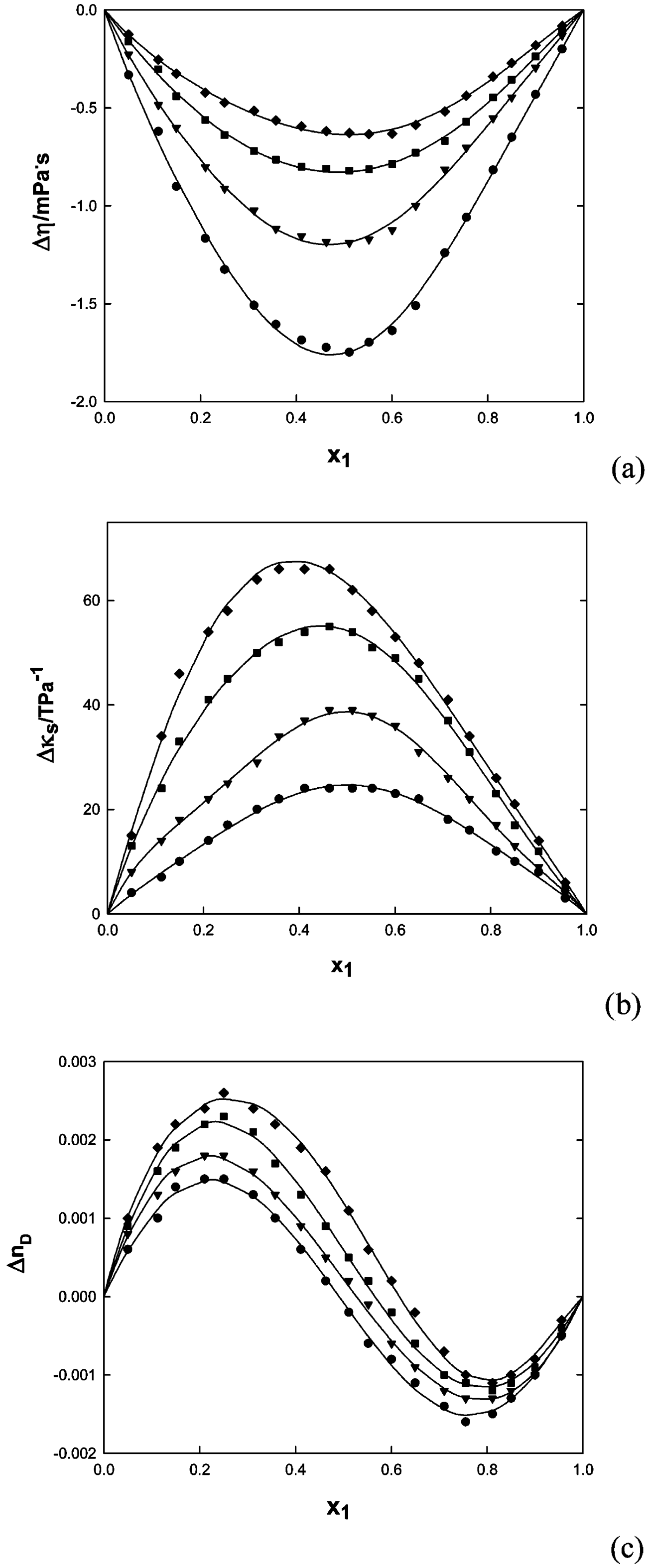

Figure 2. (a) Variation of the deviation in the viscosity $\Delta\eta$. (b) Variation of the deviation in the isentropic compressibility $\Delta\kappa_S$. (c) Variation of the deviation in the refractive index Δn_D, with mole fraction x_1 for x_1 diethylcarbonate + $(1 - x_1)$ 1-hexanol at the temperatures: ●, 293.15 K; ▼, 303.15 K; ■, 313.15 K; ◆, 323.15 K. Solid curves were calculated from the Redlich−Kister equation.

than the forces between like molecules due to differences in the shape and size of component molecules. The positive $\Delta\kappa_S$ values of binary mixtures are due to the dispersion interactions between unlike molecules, and the negative $\Delta\kappa_S$ values of the (acetophenone + 1-hexanol) binary mixture are due to the polar interactions between the hydrogen atom of the −OH group of 1-hexanol and oxygen atoms of acetophenone. The Δn_D values are positive for systems formed by (diethylcarbonate + ac-

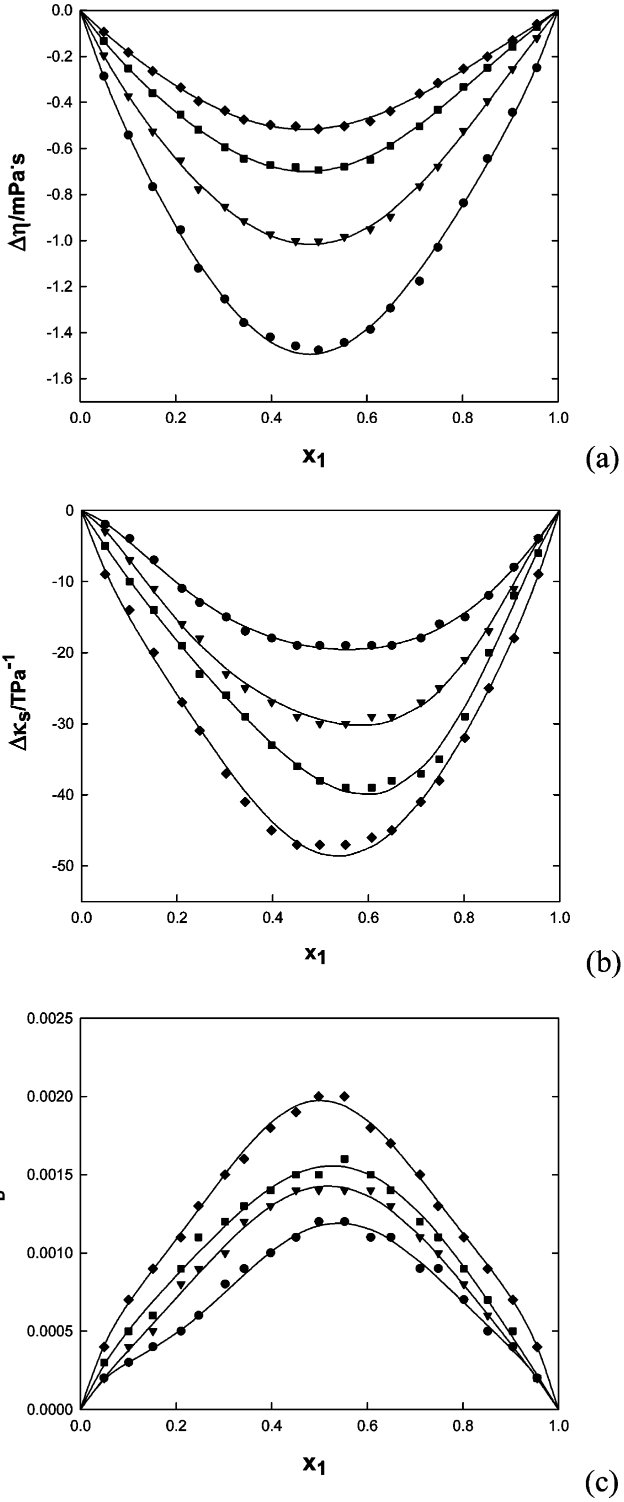

Figure 3. (a) Variation of the deviation in the viscosity $\Delta\eta$. (b) Variation of the deviation in the isentropic compressibility $\Delta\kappa_S$. (c) Variation of the deviation in the refractive index Δn_D, with mole fraction x_1 for x_1 acetophenone + $(1 - x_1)$ 1-hexanol at the temperatures: ●, 293.15 K; ▼, 303.15 K; ■, 313.15 K; ◆, 323.15 K. Solid curves were calculated from the Redlich−Kister equation.

etophenone) and (acetophenone + 1-hexanol) and become more positive with increasing temperature from (293.15 to 323.15) K. The deviations in the refractive index of (diethylcarbonate + 1-hexanol) show an S-shaped dependence on composition with positive values in the 1-hexanol-rich region and negative values at the opposite extreme. Positive values of deviations in the refractive index, Δn_D (Figures 1 and 3c), and with an initial positive region (Figure 2c) show that dispersion forces in the mixture are higher than in the pure liquids. The negative values of Δn_D at high diethylcarbonate mole fraction for the system of (diethylcarbonate + 1-hexanol) could be explained by considering that when the packing effect decreases the number of dipoles per unit volume diminishes and therefore n_D also becomes smaller, originating negative Δn_D.

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