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Density, Viscosity, Refractive Index, and Speed of Sound in the Binary Mixtures of 1,4-Dioxane + Ethyl Acetoacetate, + Diethyl Oxalate, + Diethyl Phthalate, or + Dioctyl Phthalate at 298.15, 303.15, and 308.15 K

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Experimental results of density, viscosity, and refractive index at (298.15, 303.15, and 308.15) K and speed of sound at 298.15 K are measured for the binary mixtures of 1,4-dioxane with ethyl acetoacetate, diethyl oxalate, diethyl phthalate, and dioctyl phthalate over the entire range of mixture composition. From these data, excess molar volume, deviations in viscosity, molar refraction, speed of sound, and isentropic compressibility have been calculated. These results are fitted to the polynomial equation to derive the coefficients and standard errors. The experimental and calculated quantities are used to study the nature of mixing behaviors between mixture components.

Introduction

Having wide usage in flavoring, perfumery, artificial essences, and cosmetics, esters become one of the industrially important classes of liquids. Esters are also used as solvents in pharmaceutical and paint industries and as plasticizers in plastic industries. Among different type of esters, ethyl acetoacetate, diethyl phthalate, diethyl oxalate, and dioctyl phthalate are commonly used.

In a previous paper,[1] we have presented binary mixture data on mixtures of 1,4-dioxane with alkylamines, alkanes, and an alkyldiol. In continuation of earlier work,[2−10] on mixtures containing 1,4-dioxane and those containing esters, and as a part of our ongoing effort on the accumulation of new experimental data, we now present the results of ρ, η, n_D, and u on the binary mixtures of 1,4-dioxane with esters, viz., ethyl acetoacetate, diethyl phthalate, diethyl oxalate, and dioctyl phthalate, measured at different temperatures over the entire mixture composition. To the best of our knowledge, such data on the above-mentioned mixtures are not available in the earlier literature. From the experimental data of ρ, η, n_D, and u, excess molar volume (V^E), deviation in viscosity ($\Delta\eta$), deviation in Lorentz−Lorentz molar refractivity (ΔR), deviation in speed of sound (Δu), and deviation in isentropic compressibility (Δk_s) have been computed. These data are discussed to study the nature of behaviors between the components of the mixtures, the study of which will have great relevance in chemical process engineering.

Experimental Section

Materials. High purity laboratory reagent grade samples of ethyl acetoacetate, diethyl oxalate, diethyl phthalate, and dioctyl phthalate were procured from s.d. fine Chemicals, Mumbai, India. 1,4-Dioxane is a high purity sample purchased from s.d. fine Chemicals, Mumbai, India. The mole percent purities of these liquids as determined by GC (HP 6890) using a FID detector were >99 and are reported in Table 1 along with the comparison of density and refractive index data measured at 298.15 K for pure liquids with the literature values.

Table 1. Comparison of Experimental Densities (ρ) and Refractive Indices (n_D) of Pure Liquids with Literature Values at 298.15 K

liquid	mol % purity	ρ/kg·m^{-3} expt	ρ/kg·m^{-3} lit.	n_D expt	n_D lit.
1,4-dioxane	>99.0	1027.6	1028.0[2]	1.4201	1.4203[2]
ethyl acetoacetate	>99.0	1020.8	1021.2[22]	1.4189	1.4192[23] [a]
diethyl oxalate	>99.5	1078.7	1078.5[24] [a]	1.4107	1.4101[24] [a]
diethyl phthalate	>97.0	1119.6	1117.5[24] [a]	1.5024	1.5019[24] [a]
dioctyl phthalate	>99.7	980.6	978.0[23]	1.4850	1.4848[23]

[a] Measured and compared at 293.15 K.

Binary mixtures were prepared by mass[11] in specially designed airtight glass bottles. The mass measurements accurate to ±0.01 mg were performed on a digital electronic balance (Mettler, AE240, Switzerland). A set of nine compositions were prepared for each mixture, and their physical properties were measured at the respective compositions in the mole fraction scale from 0.1 to 0.9 in steps of 0.1. In all cases, the possible error in mole fraction was less than 0.0002.

Methods. Densities of liquids and liquid mixtures were measured with an accuracy of ±0.0001 g·cm^{-3} using the capillary-type pycnometer of capacity 10 cm^3. The experimental details of density measurements and calibrations of the pycnometer are the same as those reported earlier.[11−13]

Viscosities were measured using a Cannon Fenske viscometer (size 75, ASTM D 445, Industrial Research Glassware Ltd., Roselle, NJ). An electronic digital stopwatch with a readability of ±0.01 s was used for the flow time measurements. The measured viscosity values are accurate to ±0.001 mPa·s. The calibration procedures of the pycnometer remain the same as those described previously.[11,12]

Refractive indices for the sodium D-line were measured using a thermostatically controlled Abbe refractometer

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Table 2. Experimental Density (ρ), Viscosity (η), Refractive Index (n_D), and Speed of Sound (u) of the Binary Mixtures at Different Temperatures

x_1	ρ/kg·m^{-3}	η/mPa·s	n_D	u/m·s^{-1}	x_1	ρ/kg·m^{-3}	η/mPa·s	n_D	u/m·s^{-1}
1,4-Dioxane (1) + Ethyl Acetoacetate (2)					1,4-Dioxane (1) + Diethyl Phthalate (2)				
		298.15 K					298.15 K		
0.0	1020.9	1.480	1.4166	1331	0.0	1115.1	7.926	1.5001	1446
0.1134	1021.5	1.425	1.4170	1335	0.1133	1110.7	6.520	1.4962	1435
0.2058	1022.1	1.386	1.4171	1338	0.2083	1106.5	5.536	1.4924	1427
0.3054	1022.7	1.346	1.4174	1340	0.3086	1101.4	4.593	1.4882	1418
0.4008	1023.3	1.309	1.4176	1342	0.4090	1095.5	3.793	1.4826	1408
0.4986	1024.0	1.272	1.4178	1344	0.5026	1089.1	3.122	1.4768	1399
0.5955	1024.7	1.237	1.4180	1345	0.5985	1081.4	2.544	1.4700	1390
0.6972	1025.4	1.208	1.4186	1346	0.7020	1071.6	2.037	1.4608	1380
0.7971	1026.1	1.182	1.4192	1346	0.8001	1060.2	1.641	1.4504	1370
0.8964	1026.8	1.166	1.4195	1347	0.8975	1046.3	1.319	1.4376	1359
1.0	1027.6	1.166	1.4201	1347	1.0	1027.6	1.167	1.4202	1347
		303.15 K					303.15 K		
0.0	1014.9	1.339	1.4145		0.0	1110.2	6.129	1.4978	
0.1134	1016.0	1.311	1.4146		0.1133	1105.8	5.327	1.4940	
0.2058	1016.5	1.276	1.4148		0.2083	1101.5	4.567	1.4902	
0.3054	1017.1	1.240	1.4150		0.3086	1096.3	3.813	1.4860	
0.4008	1017.6	1.205	1.4154		0.4090	1090.4	3.180	1.4805	
0.4986	1018.2	1.172	1.4155		0.5026	1084.0	2.637	1.4747	
0.5955	1018.8	1.140	1.4160		0.5985	1076.1	2.184	1.4676	
0.6972	1019.5	1.115	1.4162		0.7020	1066.1	1.753	1.4585	
0.7971	1020.3	1.091	1.4166		0.8001	1054.7	1.436	1.4481	
0.8964	1021.0	1.077	1.4169		0.8975	1040.7	1.180	1.4350	
1.0	1021.9	1.069	1.4170		1.0	1021.9	1.069	1.4170	
		308.15 K					308.15 K		
0.0	1010.2	1.239	1.4120		0.0	1105.6	4.893	1.4958	
0.1134	1010.6	1.197	1.4122		0.1133	1101.0	4.134	1.4917	
0.2058	1011.0	1.165	1.4124		0.2083	1096.6	3.597	1.4882	
0.3054	1011.4	1.133	1.4126		0.3086	1091.4	3.032	1.4837	
0.4008	1011.9	1.102	1.4130		0.4090	1085.3	2.568	1.4783	
0.4986	1012.4	1.072	1.4130		0.5026	1078.7	2.151	1.4724	
0.5955	1013.0	1.044	1.4132		0.5985	1070.9	1.824	1.4653	
0.6972	1013.7	1.021	1.4137		0.7020	1060.8	1.468	1.4564	
0.7971	1014.4	1.000	1.4140		0.8001	1049.2	1.230	1.4457	
0.8964	1015.2	0.988	1.4144		0.8975	1035.0	1.042	1.4326	
1.0	1016.2	0.984	1.4144		1.0	1016.2	0.984	1.4144	
1,4-Dioxane (1) + Diethyl Oxalate (2)					1,4-Dioxane (1) + Dioctyl Phthalate (2)				
		298.15 K					298.15 K		
0.0	1072.9	1.759	1.4084	1276	0.0	980.7	43.03	1.4850	1421
0.1094	1070.9	1.690	1.4092	1284	0.1337	982.2	32.58	1.4836	1416
0.2069	1067.5	1.631	1.4101	1292	0.2243	983.4	26.39	1.4822	1412
0.3098	1064.4	1.566	1.4112	1301	0.3207	984.8	20.73	1.4812	1406
0.4009	1061.1	1.505	1.4119	1310	0.4226	986.5	15.40	1.4781	1400
0.4975	1057.3	1.440	1.4129	1320	0.5169	988.4	11.02	1.4733	1394
0.5978	1052.8	1.375	1.4144	1329	0.6054	990.7	8.056	1.4692	1388
0.7008	1047.7	1.312	1.4157	1337	0.7161	995.0	4.947	1.4630	1379
0.7971	1042.1	1.259	1.4170	1343	0.8031	1000.2	3.213	1.4566	1372
0.8976	1035.5	1.206	1.4183	1347	0.9017	1009.9	1.863	1.4422	1361
1.0	1027.6	1.167	1.4202	1348	1.0	1027.6	1.166	1.4202	1348
		303.15 K					303.15 K		
0.0	1066.2	1.581	1.4060		0.0	977.1	30.18	1.4831	
0.1094	1064.2	1.539	1.4068		0.1337	978.3	24.97	1.4819	
0.2069	1061.4	1.487	1.4077		0.2243	979.4	20.41	1.4798	
0.3098	1058.3	1.428	1.4087		0.3207	980.5	16.10	1.4784	
0.4009	1055.0	1.375	1.4094		0.4226	982.2	12.12	1.4771	
0.4975	1051.1	1.316	1.4106		0.5169	983.9	8.852	1.4717	
0.5978	1046.7	1.259	1.4118		0.6054	986.3	6.528	1.4673	
0.7008	1041.5	1.203	1.4132		0.7161	990.5	4.096	1.4610	
0.7971	1036.1	1.157	1.4145		0.8031	995.4	2.717	1.4541	
0.8976	1029.5	1.111	1.4158		0.9017	1004.6	1.612	1.4401	
1.0	1021.9	1.069	1.4170		1.0	1021.9	1.069	1.4170	
		308.15 K					308.15 K		
0.0	1060.8	1.437	1.4034		0.0	973.1	21.88	1.4810	
0.1094	1058.1	1.388	1.4043		0.1337	974.4	17.36	1.4804	
0.2069	1055.3	1.343	1.4052		0.2243	975.2	14.44	1.4775	
0.3098	1052.1	1.291	1.4062		0.3207	976.4	11.47	1.4756	
0.4009	1048.9	1.245	1.4070		0.4226	977.8	8.836	1.4762	
0.4975	1045.0	1.193	1.4084		0.5169	979.6	6.689	1.4700	
0.5978	1040.6	1.143	1.4090		0.6054	981.9	5.001	1.4652	
0.7008	1035.4	1.094	1.4106		0.7161	985.9	3.246	1.4600	
0.7971	1029.9	1.055	1.4120		0.8031	990.6	2.222	1.4516	
0.8976	1023.6	1.016	1.4132		0.9017	999.5	1.361	1.4379	
1.0	1016.2	0.984	1.4144		1.0	1016.2	0.984	1.4144	

(Atago 3T, Japan). A minimum of three independent readings were taken for each composition, and their average value was considered in all the calculations. Refractive index data are accurate to ±0.0001 units.

Speed of sound was measured using a variable path single-crystal interferometer (Mittal Enterprises, model M-84, New Delhi). A crystal-controlled high-frequency generator was used to excite the transducer at a frequency of 1 MHz. Frequency was measured within an accuracy of 1 in 10^4 using a digital frequency meter. The interferometer cell was filled with the test liquid, and water was circulated around the measuring cell from a constant-temperature bath maintained at (298.15 ± 0.01) K. Details of the speed of sound measurements have been given earlier,[14,15] and these data are accurate to ±2 in 1000 $\text{m}\cdot\text{s}^{-1}$.

In all the property measurements, temperature was controlled within an accuracy of ±0.01 K using a constant-temperature bath. A Julabo immersion cooler (FT 200, Julabo Labortechnik, Gmph, Germany) was used to cool the water bath. This unit was installed at the intake of a heating circulator to draw the heat away from the circulating bath liquid. The immersion probe was connected to the instrument with a flexible and insulated tube. To prevent the immersion probe from icing, it was completely immersed in the bath liquid.

At least three independent readings of all the physical property measurements were taken for each composition, and the averages of these values are presented in Table 2.

Results and Discussion

From the density results, excess molar volumes (V^E) have been calculated as

$$V^{E} = V_{m} - V_{1}x_{1} - V_{2}x_{2} \tag{1}$$

Here, V_m is the molar volume of the mixture and V_1 and V_2 are the molar volumes of the pure components; x_i represents the mole fraction of the ith component of the mixture. The results of excess molar volume are reproducible to $\pm 0.005 \times 10^{-6}\ \text{m}^3\cdot\text{mol}^{-1}$. In a similar manner, the results of $\Delta\eta$, ΔR, and Δu have been calculated using the values of η, n_D, and u from the general relationship used earlier:[14−18]

$$\Delta Y = Y_{m} - Y_{1}x_{1} - Y_{2}x_{2} \tag{2}$$

In the above equation, ΔY represents $\Delta\eta$, ΔR, Δu, and Δk_s, respectively, while Y_m represents the respective mixture properties, viz., viscosity (η), molar refractivity (R) (calculated from the Lorenz−Lorentz relation), speed of sound (u), and k_s ($=1/u^2\rho$), isentropic compressibility, of the binary mixture; the symbol Y_i refers to the same properties for the pure components of the mixture. While calculating ΔR and Δk_s, the volume fraction, ϕ_i ($=\sum x_i v_i/\sum_{i=1}^{2} x_i v_i$), was used,[13,17,19−20] but, for calculating $\Delta\eta$ and Δu, the mole fraction, x_i, was used.

All the quantities (V^E, $\Delta\eta$, ΔR, Δu, and Δk_s) have been fitted to a Redlich and Kister equation[21] by the method of least-squares using the Marquardt algorithm,[22] to derive the binary coefficients (A_j) and standard deviation (σ)

$$V^{E}(\Delta Y) = x_{1}x_{2}\sum_{j=1}^{k} A_{j}(x_{2} - x_{1})^{j-1} \tag{3}$$

Table 3. Estimated Parameters of Eq 3 for Various Functions of the Binary Mixtures at Different Temperatures

function	temp/K	A_1	A_2	A_3	σ
1,4-Dioxane (1) + Ethyl Acetoacetate (2)					
$V^E/(10^{-6}\ \text{m}^3\cdot\text{mol}^{-1})$	298.15	−0.165	0.036	0.032	0.0009
	303.15	−0.194	−0.024	−0.275	0.0142
	308.15	0.067	0.018	0.0019	0.0005
$\Delta\eta/\text{mPa}\cdot\text{s}$	298.15	−0.207	0.098	−0.100	0.0016
	303.15	−0.135	0.130	0.041	0.0015
	308.15	−0.160	0.073	−0.041	0.0013
$\Delta R/10^6\ (\text{m}^3\cdot\text{mol}^{-1})$	298.15	−4.147	0.816	−0.043	0.0070
	303.15	−4.088	0.973	−0.278	0.0105
	308.15	−4.010	0.855	−0.038	0.0061
$\Delta u/\text{m}\cdot\text{s}^{-1}$	298.15	17.35	4.56	−2.36	0.124
$\Delta k_S/\text{TPa}^{-1}$	298.15	−19.13	10.51	−2.20	0.127
1,4-Dioxane (1) + Diethyl Oxalate (2)					
$V^E/(10^{-6}\ \text{m}^3\cdot\text{mol}^{-1})$	298.15	−0.744	0.078	−0.063	0.0045
	303.15	−0.812	−0.281	−0.392	0.0162
	308.15	−0.553	−0.034	−0.010	0.0033
$\Delta\eta/\text{mPa}\cdot\text{s}$	298.15	−0.094	0.126	−0.044	0.0021
	303.15	−0.040	0.156	0.084	0.0009
	308.15	−0.073	0.110	0.001	0.0016
$\Delta R/10^6\ (\text{m}^3\cdot\text{mol}^{-1})$	298.15	−5.557	1.334	−0.393	0.0116
	303.15	−5.523	1.498	−0.446	0.0137
	308.15	−5.431	1.379	−0.243	0.0169
$\Delta u/\text{m}\cdot\text{s}^{-1}$	298.15	29.59	−48.83	9.34	0.264
$\Delta k_S/\text{TPa}^{-1}$	298.15	−66.15	−16.12	14.13	0.402
1,4-Dioxane (1) + Diethyl Phthalate (2)					
$V^E/(10^{-6}\ \text{m}^3\cdot\text{mol}^{-1})$	298.15	−0.253	0.001	0.007	0.0014
	303.15	−0.261	−0.097	−0.061	0.0057
	308.15	−0.167	−0.007	0.038	0.0010
$\Delta\eta/\text{mPa}\cdot\text{s}$	298.15	−5.587	−0.402	−0.620	0.0205
	303.15	−3.785	0.837	0.675	0.0374
	308.15	−3.045	0.291	−0.467	0.0157
$\Delta R/10^6\ (\text{m}^3\cdot\text{mol}^{-1})$	298.15	−29.00	12.11	−5.501	0.0477
	303.15	−28.89	12.10	−5.428	0.0432
	308.15	−28.82	12.09	−5.462	0.0592
$\Delta u/\text{m}\cdot\text{s}^{-1}$	298.15	10.43	−6.24	2.90	0.138
$\Delta k_S/\text{TPa}^{-1}$	298.15	30.18	−14.91	5.12	0.073
1,4-Dioxane (1) + Dioctyl Phthalate (2)					
$V^E/(10^{-6}\ \text{m}^3\cdot\text{mol}^{-1})$	298.15	0.971	−1.943	0.452	0.0210
	303.15	1.432	−1.399	0.626	0.0199
	308.15	1.522	−1.426	0.335	0.0147
$\Delta\eta/\text{mPa}\cdot\text{s}$	298.15	−40.86	−1.684	1.356	0.1861
	303.15	−24.87	7.258	10.04	0.1732
	308.15	−17.54	1.986	1.409	0.0818
$\Delta R/10^6\ (\text{m}^3\cdot\text{mol}^{-1})$	298.15	−114.9	79.08	−96.73	0.7837
	303.15	−114.8	78.96	−95.53	0.8575
	308.15	−114.6	78.51	−95.19	0.9167
$\Delta u/\text{m}\cdot\text{s}^{-1}$	298.15	43.35	−17.43	23.18	0.068
$\Delta k_S/\text{TPa}^{-1}$	298.15	46.7	−23.6	3.9	0.603

In each case, the optimum number of coefficients (A_j) was determined from an examination of the variation of standard deviation (σ) as calculated by

$$\sigma = \left(\sum (Y_{\text{cal}}^{E} - Y_{\text{obs}}^{E})^{2}/(n - m)\right)^{1/2} \tag{4}$$

where n represents the number of measurements and m is the number of coefficients used in fitting the data. Estimated values of A_j and σ for V^E, $\Delta\eta$, ΔR, Δu, and Δk_s are given in Table 3. In all cases, the best fit was found by using only three adjustable fitting coefficients in eq 3.

The variations of excess molar volume (V^E) with mole fraction (x_1) of 1,4-dioxane at 298.15 K for the binary mixtures of 1,4-dioxane with ethyl acetoacetate, diethyl phthalate, diethyl oxalate, or dioctyl phthalate are displayed in Figure 1. For the mixtures of 1,4-dioxane with dioctyl phthalate, we find that, up to $x_1 = 0.2$, the V^E values are almost zero, while, in the remaining composition range, the positive values of V^E are observed with a maximum value of V^E observed at $x_1 \approx 0.7$. Such a skewed behavior is somewhat uncommon in the literature. This behavior may be attributed to the steric hindrance due to the bulky groups present in the molecule. However, with the remain-

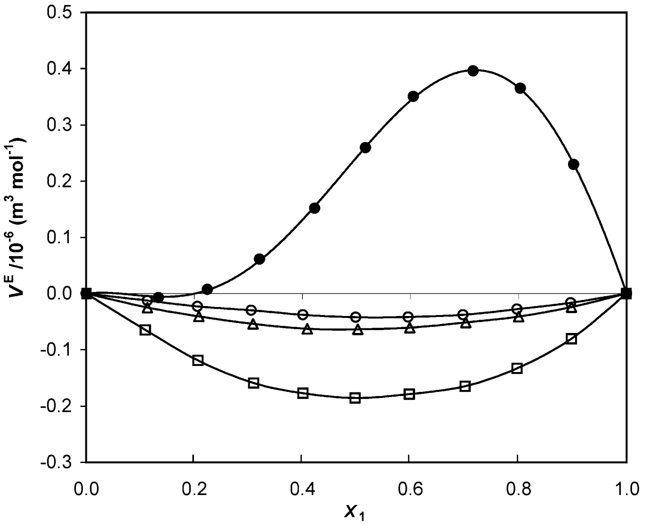

Figure 1. Excess molar volume vs mole fraction of 1,4-dioxane with (○) ethyl acetoacetate, (□) diethyl oxalate, (△) diethyl phthalate, and (●) dioctyl phthalate at 298.15 K.

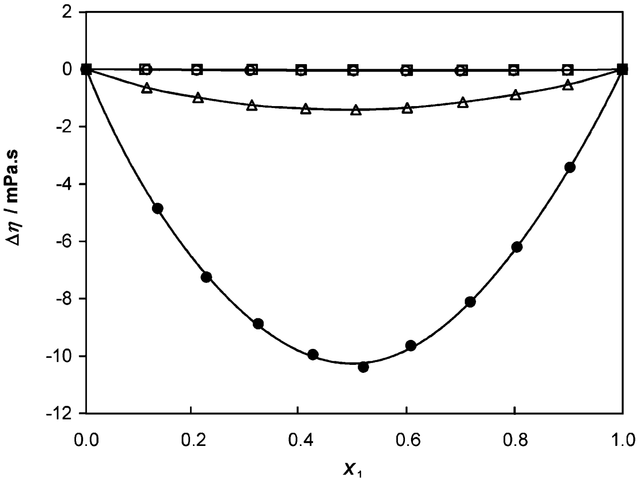

Figure 2. Deviations in viscosity vs mole fraction at 298.15 K for the same mixtures presented in Figure 1.

ing binary mixtures, V^E values are found to be negative over the entire composition range of the mixtures, indicating the specific interactions such as intermolecular hydrogen bonding between the component liquids and also the interstitial accommodation of the component liquids because of the difference in their molar volumes. The negative values may be due to the difference in the dielectric constants of the liquid components of the mixtures. The dielectric constants of 1,4-dioxane, ethyl acetoacetate, diethyl oxalate, and dioctyl phthalate are 2.21, 15.7, 8.2, and 5.1, respectively. The negative V^E values of these mixtures vary in the order diethyl oxalate > diethyl phthalate > ethyl acetoacetate. This is in accordance with the systematic variation with the change in polarity of the liquid components of the mixtures.

The results of $\Delta\eta$ versus x_1 displayed in Figure 2 at 298.15 K indicate large negative values for 1,4-dioxane + dioctyl phthalate mixtures, whereas small negative values of $\Delta\eta$ are observed for mixtures of 1,4-dioxane + diethyl phthalate. On the other hand, for mixtures containing diethyl oxalate or ethyl acetoacetate, the curves of $\Delta\eta$ versus x_1 are quite identical, and hence, a single common curve is displayed for these mixtures.

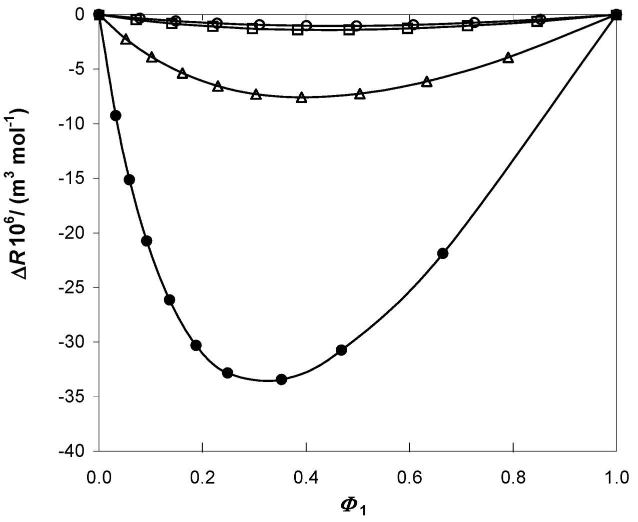

Figure 3. Deviations in molar refraction (ΔR) vs volume fraction at 298.15 K for the same mixtures presented in Figure 1.

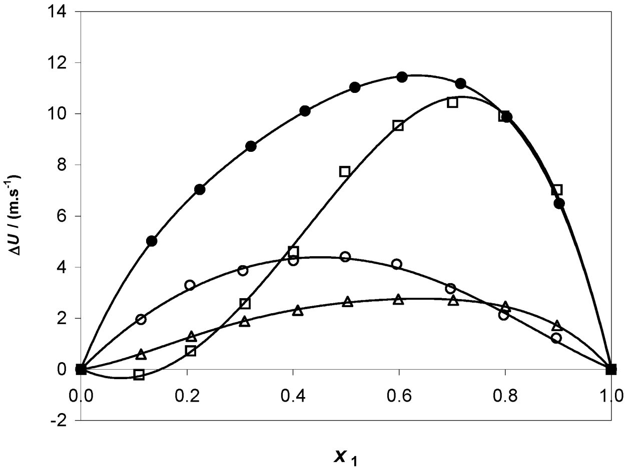

Figure 4. Deviations in speed of sound (Δu) vs mole fraction at 298.15 K for the same mixtures presented in Figure 1.

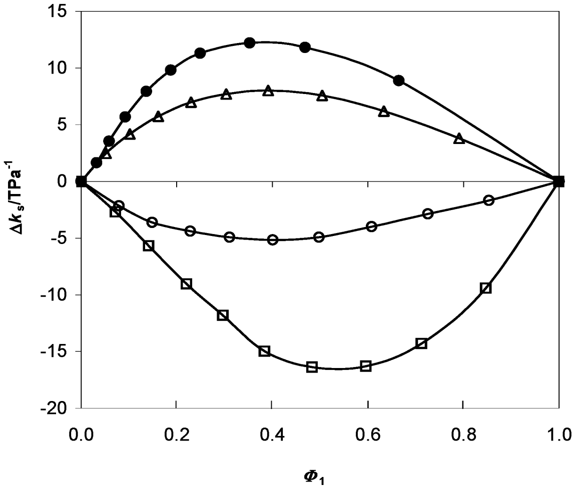

Figure 5. Deviations in isentropic compressibility (Δk_s) vs volume fraction at 298.15 K for the same mixtures presented in Figure 1.

The dependence of molar refraction (ΔR) on volume fraction (ϕ_1) of 1,4-dioxane at 298.15 K is presented in Figure 3. It is observed that, for the mixture of 1,4-dioxane with dioctyl phthalate, ΔR is large and negative over the whole of the mixture composition, whereas, for 1,4-dioxane

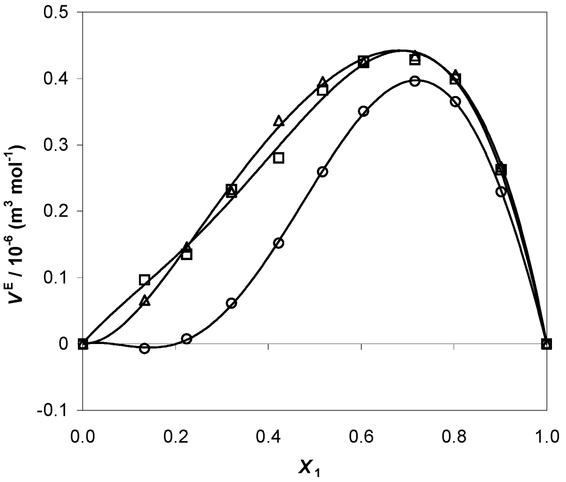

Figure 6. Effect of temperature on V^E for the 1,4-dioxane + dioctyl phthalate mixture at (○) 298.15 K, (□) 303.15 K, and (△) 308.15 K.

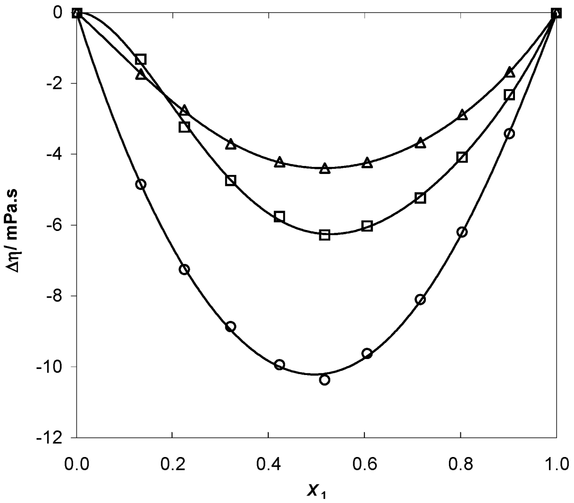

Figure 7. Effect of temperature on $\Delta\eta$ for the 1,4-dioxane + dioctyl phthalate mixture at (○) 298.15 K, (□) 303.15 K, and (△) 308.15 K.

+ diethyl phthalate mixtures, the ΔR values are small but negative. However, in the case of mixtures of 1,4-dioxane + ethyl acetoacetate or + diethyl oxalate, the values of ΔR are slightly negative and the curves vary almost identically over the entire mole fraction scale; hence, a single curve is drawn to display both their dependencies.

The dependence of deviations in speed of sound (Δu) on mole fraction of 1,4-dioxane (x_1) is presented in Figure 4. For all binary mixtures, the values of Δu are positive except in the case of the 1,4-dioxane + diethyl oxalate mixture, for which the curve was skewed before $x_1 = 0.2$, with slight negative values observed at $x_1 = 0.1$. Also, the maximum values of Δu for 1,4-dioxane + dioctyl phthalate or + diethyl oxalate are slightly shifted toward the higher mixture composition scale (i.e., at around x_1= 0.7). In the case of 1,4-dioxane + ethyl acetoacetate, Δu values vary systematically over the entire mole fraction scale.

The results of Δk_s versus ϕ_1 for all the mixtures at 298.15 K are presented in Figure 5. It is observed that, for the mixtures of 1,4-dioxane with diethyl oxalate or ethyl acetoacetate, the Δk_s values are negative; for the latter mixture, the values are higher than those for the former. For mixtures of 1,4-dioxane with diethyl phthalate or dioctyl phthalate, the Δk_s values are positive and highest for the latter.

The effect of temperature on excess molar volume (V^E) and deviation in viscosity ($\Delta\eta$) is quite systematic, as displayed typically in Figures 6 and 7 for the dioxane + dioctyl phthalate mixture. It is found that V^E results become more positive with increasing temperature whereas $\Delta\eta$ versus x_1 plots exhibit similar dependencies on temperature but their magnitude decreases systematically with increasing temperature and the values are negative.

It may be noted that, in all the plots, points represent the quantities calculated from eqs 1 and 2 while the smooth curves are drawn from the best-fitted values calculated from eq 3.

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