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Density, Viscosity, Refractive Index, and Speed of Sound in the Binary Mixtures of Ethyl Chloroacetate with Aromatic Liquids at 298.15, 303.15, and 308.15 K

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Densities, viscosities, and refractive indices at 298.15, 303.15, and 308.15 K and speed of sound values at 298.15 K have been presented for the binary mixtures of ethyl chloroacetate + benzene, toluene, 1,4-dimethylbenzene (*p*-xylene), 1,3,5-trimethylbenzene (mesitylene), and methoxybenzene (anisole). From the results of density, viscosity, and refractive index, the excess molar volume, deviations in viscosity, molar refractivity, speed of sound, and isentropic compressibility have been calculated. These results have been correlated by a polynomial Redlich−Kister polynomial equation to derive the coefficients and standard errors. Variations of these quantities have been displayed graphically.

Introduction

In our previous paper[1] we presented the thermodynamic and transport data on the binary mixtures of ethyl chloroacetate with alkanes. In a continuation of this study and as a part of our ongoing program of research,[2−5] we present here the experimental data on density, ρ, viscosity, η, and refractive index, n_D, for the sodium D line at 298.15, 303.15, and 308.15 K and speed of sound, u, at 298.15 K for the binary mixtures of ethyl chloroacetate with benzene, toluene, *p*-xylene, mesitylene, and anisole. Aromatic liquids used in this study are important in petrochemical industries, while ethyl chloroacetate is used as a solvent in organic reactions.[6] Therefore, measurement of physicochemical property data on such mixtures will be useful in process engineering. We are not aware of any such data in the earlier literature on the mixtures of this study and, hence, an attempt has been made to present the data on ρ, η, n_D, and u and to compute the excess molar volume, V^E, and deviations in viscosity, $\Delta\eta$, molar refraction, ΔR, speed of sound, Δu, and isentropic compressibility, Δk_s, for the mixtures of ethyl chloroacetate + benzene, toluene, *p*-xylene, mesitylene, or anisole over the entire range of mixture composition. These results have been fitted to the Redlich−Kister equation[7] to derive the binary coefficients and to estimate the standard errors. The results are displayed graphically.

Experimental Section

Materials. A high-purity spectroscopic-grade sample of toluene was procured from Spectrochem Ltd., Mumbai, India. Benzene was obtained from Ranbaxy, Mumbai, India. *p*-Xylene and anisole were procured from s.d. fine Chemicals, Mumbai, India. Ethyl chloroacetate was purchased from BASCO, Mumbai, India. Mesitylene is an analytical reagent-grade sample, purchased from Merck. The purities of these liquids as determined by gas−liquid chromatography (HP 6890) using a flame ionization detector were >99 mol %, and these data are reported in Table 1 along with the results of density and refractive index at 298.15 K for pure liquids, which are compared 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.
ethyl chloroacetate	>99.0	1157.0[a]	1158.5[a,1]	1.4211[a]	1.4215[a,1]
benzene	>99.9	873.1	873.6[18]	1.4979	1.4979[17]
toluene	>99.7	679.3	679.5[18]	1.3859	1.3851[17]
p-xylene	>99.0	856.6	856.6[17]	1.4933	1.4932[17]
mesitylene	>99.0	861.7	861.1[17]	1.4974	1.4968[17]
anisole	>99.5	989.0	989.3[17]	1.5152	1.5143[17]

[a] Measured and compared at 293.15 K.

Binary mixtures were prepared by mass in airtight glass bottles.[8] The mass measurements, accurate to ±0.01 mg, were made on a digital electronic balance (Mettler AE 240, Switzerland). A set of nine compositions was prepared for each mixture, and their physical properties were measured at the respective composition of the mole fraction varying from 0.1 to 0.9 in steps of 0.1. In all of the cases, the possible error in the mole fraction was estimated to be less than 10^{-4}.

Methods. Densities of liquids and liquid mixtures were measured to an accuracy of ±0.0005 g·cm^{-3} using a capillary pycnometer of about 10 cm^3 volume. Experimental details on density measurements were given earlier.[8,9] Calibrations of the pycnometer have been described earlier.[8,10] Viscosities were measured using a Cannon Fenske viscometer (size 100, 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 ±0.005 mPa·s

Refractive indices for the sodium D line were measured using a thermostatically controlled Abbe refractometer (Atago 3T, Japan). A minimum of three independent readings was taken for each composition, and their average value is used in all of the calculations. The results of refractive indices are accurate to ±0.0001 units.

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

x_1	ρ/kg·cm^{-3}	η/mPa·s	n_D	u/m·s^{-1}	x_1	ρ/kg·cm^{-3}	η/mPa·s	n_D	u/m·s^{-1}
Ethyl Chloroacetate (1) + Benzene (2)									
298.15 K									
0.0000	873.1	0.610	1.4979	1304	0.6023	1048.0	0.834	1.4456	1258
0.0985	904.1	0.634	1.4880	1301	0.6960	1073.0	0.886	1.4393	1255
0.1983	934.6	0.659	1.4785	1289	0.7994	1098.0	0.949	1.4324	1253
0.2974	963.9	0.694	1.4694	1275	0.8998	1122.0	1.019	1.4260	1250
0.3925	991.2	0.734	1.4615	1267	1.0000	1145.0	1.095	1.4200	1250
0.5049	1022.0	0.781	1.4526	1261					
303.15 K									
0.0000	868.0	0.569	1.4945		0.6023	1042.0	0.779	1.4426	
0.0985	898.5	0.592	1.4844		0.6960	1066.0	0.825	1.4362	
0.1983	928.8	0.616	1.4754		0.7994	1092.0	0.883	1.4294	
0.2974	957.9	0.649	1.4666		0.8998	1116.0	0.948	1.4232	
0.3925	983.9	0.687	1.4585		1.0000	1139.0	1.013	1.4173	
0.5049	1016.0	0.731	1.4498						
308.15 K									
0.0000	861.9	0.533	1.4910		0.6023	1035.0	0.723	1.4399	
0.0985	892.6	0.549	1.4813		0.6960	1060.0	0.765	1.4336	
0.1983	922.8	0.574	1.4722		0.7994	1085.0	0.820	1.4269	
0.2974	951.8	0.605	1.4634		0.8998	1109.0	0.878	1.4209	
0.3925	978.8	0.640	1.4555		1.0000	1132.0	0.930	1.4148	
0.5049	1009.0	0.681	1.4469						
Ethyl Chloroacetate (1) + Toluene (2)									
298.15 K									
0.0000	862.0	0.565	1.4944	1309	0.6083	1035.0	0.823	1.4483	1265
0.1004	890.9	0.594	1.4866	1304	0.6963	1060.0	0.878	1.4418	1259
0.2070	921.4	0.631	1.4783	1295	0.8022	1090.0	0.945	1.4342	1255
0.2990	947.8	0.671	1.4713	1287	0.9006	1117.0	1.020	1.4272	1252
0.4013	977.0	0.714	1.4637	1279	1.0000	1145.0	1.095	1.4200	1250
0.5048	1006.0	0.768	1.4560	1271					
303.15 K									
0.0000	857.1	0.534	1.4913		0.6083	1029.0	0.770	1.4454	
0.1004	885.5	0.561	1.4833		0.6963	1054.0	0.818	1.4389	
0.2070	915.4	0.594	1.4756		0.8022	1083.0	0.881	1.4313	
0.2990	942.1	0.630	1.4682		0.9006	1110.0	0.947	1.4247	
0.4013	971.4	0.669	1.4608		1.0000	1139.0	1.013	1.4173	
0.5048	1000.0	0.719	1.4531						
308.15 K									
0.0000	850.9	0.503	1.4881		0.6083	1023.0	0.718	1.4422	
0.1004	879.9	0.528	1.4802		0.6963	1048.0	0.758	1.4360	
0.2070	910.4	0.559	1.4720		0.8022	1077.0	0.817	1.4285	
0.2990	936.7	0.590	1.4652		0.9006	1103.0	0.873	1.4220	
0.4013	965.8	0.626	1.4575		1.0000	1132.0	0.930	1.4148	
0.5048	994.8	0.671	1.4498						
Ethyl Chloroacetate (1) + *p*-Xylene (2)									
298.15 K									
0.0000	856.7	0.613	1.4933	1326	0.6010	1019.0	0.832	1.4511	1266
0.0994	881.5	0.635	1.4865	1320	0.6994	1049.0	0.886	1.4437	1259
0.1995	907.3	0.663	1.4796	1312	0.7968	1079.0	0.949	1.4363	1255
0.3008	934.0	0.699	1.4728	1301	0.8943	1110.0	1.017	1.4288	1251
0.3992	960.9	0.735	1.4659	1290	1.0000	1145.0	1.095	1.4200	1250
0.5001	989.6	0.783	1.4585	1277					
303.15 K									
0.0000	852.2	0.574	1.4904		0.6010	1013.0	0.778	1.4482	
0.0994	876.1	0.599	1.4837		0.6994	1042.0	0.827	1.4411	
0.1995	902.4	0.624	1.4770		0.7968	1073.0	0.882	1.4337	
0.3008	928.5	0.656	1.4700		0.8943	1104.0	0.945	1.4262	
0.3992	955.7	0.691	1.4631		1.0000	1139.0	1.013	1.4173	
0.5001	983.9	0.734	1.4558						
308.15 K									
0.0000	847.5	0.542	1.4875		0.6010	1007.0	0.726	1.4455	
0.0994	871.9	0.563	1.4808		0.6994	1036.0	0.768	1.4384	
0.1995	897.3	0.586	1.4741		0.7968	1066.0	0.816	1.4308	
0.3008	923.7	0.615	1.4670		0.8943	1097.0	0.874	1.4234	
0.3992	950.2	0.646	1.4603		1.0000	1132.0	0.930	1.4148	
0.5001	978.4	0.686	1.4528						
Ethyl Chloroacetate (1) + Mesitylene (2)									
298.15 K									
0.0000	861.8	0.671	1.4974	1345	0.5997	1009.0	0.853	1.4542	1276
0.1123	886.0	0.693	1.4902	1329	0.6963	1039.0	0.902	1.4464	1268
0.2035	906.8	0.712	1.4844	1318	0.7974	1072.0	0.964	1.4381	1262
0.2999	929.9	0.745	1.4768	1307	0.8954	1106.0	1.023	1.4296	1255
0.4019	955.6	0.775	1.4695	1295	1.0000	1145.0	1.095	1.4200	1250
0.4987	981.4	0.811	1.4623	1286					
303.15 K									
0.0000	856.8	0.624	1.4950		0.5997	1004.0	0.798	1.4516	
0.1123	881.1	0.653	1.4873		0.6963	1033.0	0.841	1.4438	
0.2035	900.9	0.669	1.4813		0.7974	1065.0	0.899	1.4354	
0.2999	924.8	0.698	1.4742		0.8954	1100.0	0.949	1.4269	
0.4019	950.1	0.725	1.4667		1.0000	1139.0	1.013	1.4173	
0.4987	975.6	0.759	1.4593						

Table 2. Cont'd

x_1	ρ/kg·cm^{-3}	η/mPa·s	n_D	u/m·s^{-1}	x_1	ρ/kg·cm^{-3}	η/mPa·s	n_D	u/m·s^{-1}
308.15 K									
0.0000	852.6	0.589	1.4922		0.5997	998.7	0.741	1.4490	
0.1123	876.6	0.612	1.4845		0.6963	1027.0	0.779	1.4410	
0.2035	897.1	0.627	1.4784		0.7974	1059.0	0.829	1.4328	
0.2999	919.8	0.652	1.4714		0.8954	1093.0	0.874	1.4244	
0.4019	945.1	0.677	1.4640		1.0000	1132.0	0.930	1.4148	
0.4987	970.6	0.708	1.4569						
Ethyl Chloroacetate (1) + Anisole (2)									
298.15 K									
0.0000	989.0	0.991	1.5153	1418	0.6022	1083.0	1.019	1.4575	1320
0.1012	1005.0	0.983	1.5056	1401	0.6954	1098.0	1.033	1.4488	1304
0.2027	1020.0	0.987	1.4959	1385	0.7934	1113.0	1.049	1.4396	1286
0.3000	1036.0	0.989	1.4866	1370	0.8919	1128.0	1.072	1.4303	1268
0.3936	1050.0	0.996	1.4775	1355	1.0000	1145.0	1.095	1.4200	1250
0.4982	1067.0	1.004	1.4674	1338					
303.15 K									
0.0000	984.9	0.923	1.5124		0.6022	1077.0	0.945	1.4548	
0.1012	999.3	0.915	1.5028		0.6954	1092.0	0.958	1.4462	
0.2027	1015.0	0.916	1.4930		0.7934	1106.0	0.975	1.4369	
0.3000	1030.0	0.918	1.4838		0.8919	1122.0	0.993	1.4276	
0.3936	1044.0	0.925	1.4746		1.0000	1139.0	1.013	1.4173	
0.4982	1061.0	0.932	1.4649						
308.15 K									
0.0000	978.8	0.849	1.5094		0.6022	1071.0	0.871	1.4523	
0.1012	994.3	0.847	1.4999		0.6954	1085.0	0.884	1.4435	
0.2027	1009.0	0.847	1.4902		0.7934	1100.0	0.901	1.4344	
0.3000	1024.0	0.848	1.4810		0.8919	1115.0	0.915	1.4252	
0.3936	1039.0	0.853	1.4720		1.0000	1132.0	0.930	1.4148	
0.4982	1055.0	0.861	1.4620						

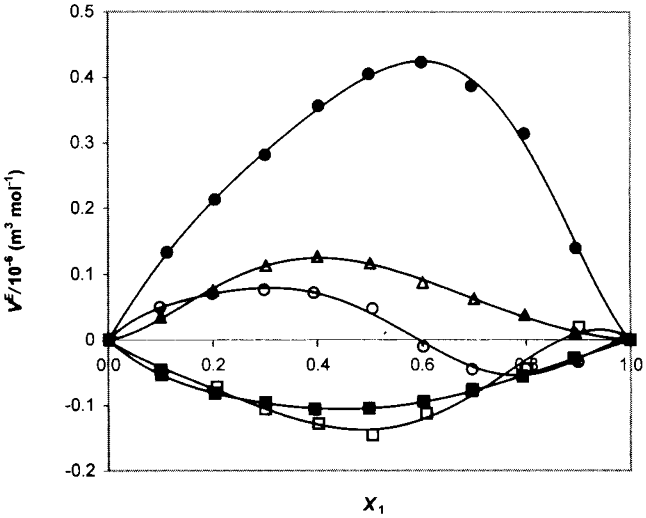

Figure 1. Excess molar volume (V^E) vs mole fraction of ethyl chloroacetate with (○) benzene, (□) toluene, (△) *p*-xylene, (●) mesitylene, and (■) anisole at 298.15 K.

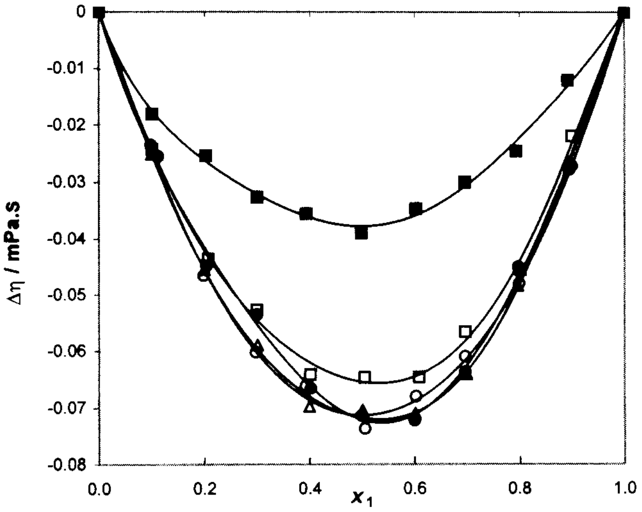

Figure 2. Deviations in viscosity ($\Delta\eta$) vs mole fraction at 298.15 K for mixtures of ethyl chloroacetate with aromatic liquids. Symbols are the same as those given in Figure 1.

The speed of sound was measured by using a variable-path single-crystal interferometer (Mittal Enterprises model M-84, New Delhi, India). A crystal-controlled high-frequency generator was used to excite the transducer at a frequency of 1 MHz. The frequency was measured within an accuracy of 1 in 10^4 using a digital frequency meter. An interferometer cell was filled with the test liquid, and water was circulated around the measuring cell from a thermostated bath maintained at 298.15 ± 0.01 K. To increase the accuracy of the measurement, several such maxima were counted by changing the distance between the transducer and reflector. The total distance, d (cm), moved by the reflector, which is fixed to the micrometer scale, corresponding to two maxima in the ammeter was measured and used to calculate the wavelength, λ (cm), by using $d = n\lambda/2$. By knowing the frequency, ν, of the crystal (1 MHz), the speed of sound, u in m·s^{-1}, was calculated as $u = \nu\lambda$. The speed of sound values thus calculated were accurate to ±2 in 1000 m·s^{-1}. The isentropic compressibilities were calculated using $k_s = 1/u^2\rho$ (where u is in m·s^{-1} and ρ in kg/m^3).

In all of the property measurements, the temperature was controlled within ±0.01 K using a constant-temperature bath. An immersion cooler (Julabo Labortechnik Gmbh FT 200, 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. 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.

For all of the properties, a set of three readings was taken for each composition, but only the average of these values is given in Table 2.

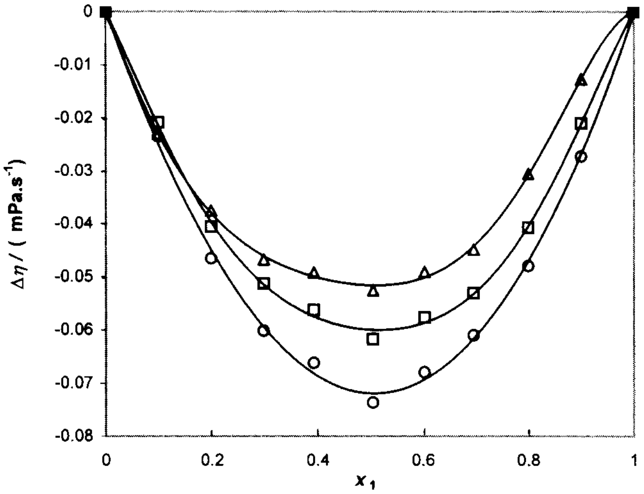

Figure 3. Effect of temperature on $\Delta\eta$ for an ethyl chloroacetate + benzene mixture: (○) 298.15 K; (□) 303.15 K; (△) 308.15 K.

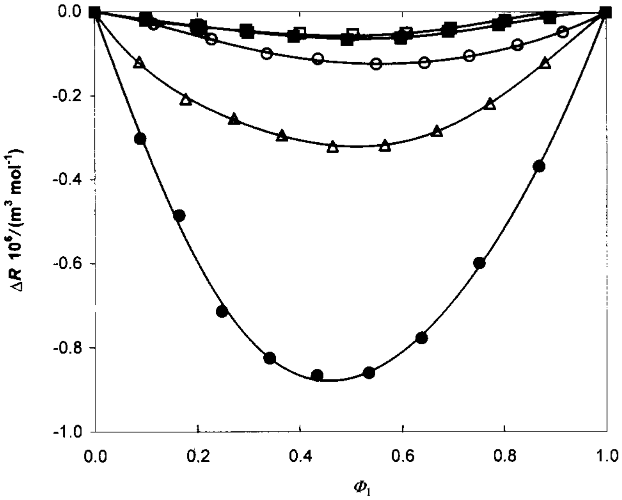

Figure 4. Deviations in molar refraction (ΔR) vs volume fraction at 298.15 K for mixtures of ethyl chloroacetate with aromatic liquids. Symbols are the same as those given in Figure 1.

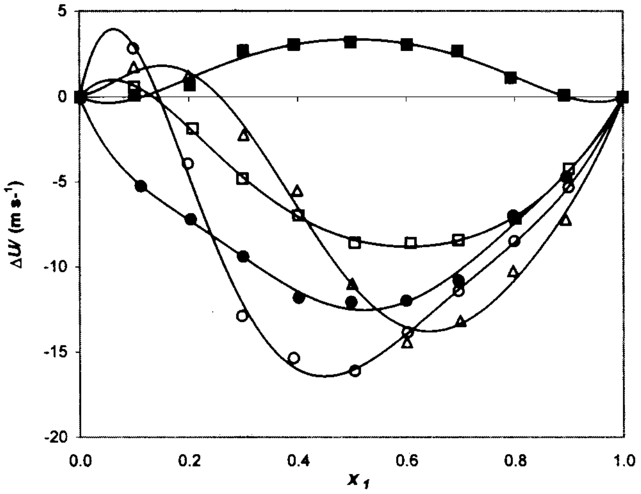

Figure 5. Deviations in the speed of sound (Δu) vs mole fraction for mixtures of ethyl chloroacetate with aromatic liquids. Symbols are the same as those given in Figure 1.

Results and Discussion

From the density results, the excess molar volume, V^E,

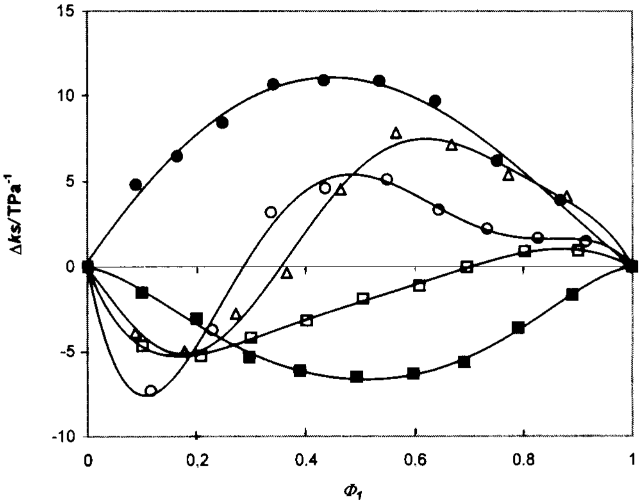

Figure 6. Deviations in the isentropic compressibility (Δk_S) vs volume fraction for mixtures of ethyl chloroacetate with aromatic liquids. Symbols are the same as those given in Figure 1.

has been calculated as

$$V^E = V_m - V_1x_1 - V_2x_2 \tag{1}$$

Here, V_m is the molar volume of the mixture, V_1 and V_2 are molar volumes of pure components, and x_i represents the mole fraction of the ith component of the mixture. From viscosity results, the deviation in the viscosity was calculated as

$$\Delta\eta = \eta_m - \eta_1x_1 - \eta_2x_2 \tag{2}$$

where η_1, η_2, and η_m are respectively the viscosities of pure components (1 and 2) of the mixtures. The refractive index values have been used to calculate the Lorentz–Lorentz molar refraction and deviations in the molar refraction have been calculated as

$$\Delta R = R_m - R_1\phi_1 - R_2\phi_2 \tag{3}$$

Here R_m is the molar refractivity of mixtures; R_1 and R_2 are the molar refractivities of pure components 1 and 2; ϕ_i represents the volume fraction of the ith component as explained before.[1,5,8,11–13]

The results of the excess molar volume at 298.15 K in Figure 1 show variations depending upon the nature of the aromatic liquid in the mixture. For instance, a large positive deviation is observed in the case of ethyl chloroacetate + mesitylene, while negative values of V^E are observed for ethyl chloroacetate + toluene or anisole. It may be noted that binary mixtures of ethyl chloroacetate + toluene or anisole exhibit almost identical trends over the whole of the mixture composition, probably because of the identical nature of the molecules. For ethyl chloroacetate + *p*-xylene, the V^E values are small but positive. For ethyl chloroacetate + benzene, a sigmoidal trend with positive and negative V^E values is observed. Such trends have also been observed earlier.[1,5,8,11–14]

The V^E results at other temperatures follow the same trends with slight variations in magnitude, but these are not displayed to avoid a redundancy of graphical presentations.

From the plots of $\Delta\eta$ vs x_1 at 298.15 K presented in Figure 2, the behavior of almost all of the mixtures is identical except ethyl chloroacetate + anisole. In general, negative values of $\Delta\eta$ vary as per the sequence anisole >

Table 3. Estimated Parameters of Equation 4 for Various Functions of the Binary Mixtures at Different Temperatures

function	temp/K	A_1	A_2	A_3	σ
Ethyl Chloroacetate (1) + Benzene (2)					
$V^E \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	0.139	0.657	−0.164	0.015
	303.15	0.419	0.539	0.502	0.049
	308.15	0.147	0.607	−0.041	0.014
$\Delta\eta/\text{mPa}\cdot\text{s}$	298.15	−0.287	0.009	−0.009	0.002
	303.15	−0.244	0.002	−0.008	0.002
	308.15	−0.211	−0.036	−0.001	0.003
$\Delta R \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	−0.486	−0.158	0.085	0.005
	303.15	−0.369	−0.191	0.072	0.017
	308.15	−0.459	−0.170	0.108	0.004
$\Delta u/\text{m}\cdot\text{s}^{-1}$	298.15	−65.77	16.17	73.62	3.044
$\Delta k_S/\text{T}\cdot\text{Pa}^{-1}$	298.15	20.68	29.20	−80.04	2.297
Ethyl Chloroacetate (1) + Toluene (2)					
$V^E \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	−0.548	−0.216	0.602	0.015
	303.15	−0.350	−0.365	1.636	0.036
	308.15	−0.888	−0.469	0.604	0.018
$\Delta\eta/\text{mPa}\cdot\text{s}$	298.15	−0.264	0.010	−0.008	0.002
	303.15	−0.232	−0.002	−0.001	0.001
	308.15	−0.196	−0.012	0.011	0.002
$\Delta R \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	−0.228	0.084	0.177	0.004
	303.15	−0.194	0.132	0.490	0.025
	308.15	−0.434	0.165	0.246	0.011
$\Delta u/\text{m}\cdot\text{s}^{-1}$	298.15	−33.85	26.44	18.44	0.561
$\Delta k_S/\text{T}\cdot\text{Pa}^{-1}$	298.15	−7.96	30.23	−17.18	0.772
Ethyl Chloroacetate (1) + *p*-Xylene (2)					
$V^E \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	0.466	0.244	−0.328	0.009
	303.15	0.588	0.186	0.762	0.058
	308.15	0.529	0.240	−0.169	0.004
$\Delta\eta/\text{mPa}\cdot\text{s}$	298.15	−0.291	0.015	0.002	0.002
	303.15	−0.247	0.018	0.017	0.001
	308.15	−0.211	−0.002	0.009	0.003
$\Delta R \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	−1.277	0.105	−0.047	0.009
	303.15	−1.229	0.103	0.401	0.018
	308.15	−1.259	0.060	0.017	0.007
$\Delta u/\text{m}\cdot\text{s}^{-1}$	298.15	−42.50	64.38	31.68	1.421
$\Delta k_S/\text{T}\cdot\text{Pa}^{-1}$	298.15	21.04	53.61	−49.35	1.032
Ethyl Chloroacetate (1) + Mesitylene (2)					
$V^E \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	1.631	−0.471	−0.191	0.036
	303.15	1.805	−0.482	0.033	0.058
	308.15	1.615	−0.454	−0.423	0.035
$\Delta\eta/\text{mPa}\cdot\text{s}$	298.15	−0.287	0.029	0.025	0.003
	303.15	−0.239	0.040	0.067	0.003
	308.15	−0.212	0.026	0.040	0.003
$\Delta R \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	−3.501	0.448	0.132	0.026
	303.15	−3.513	0.600	−0.033	0.018
	308.15	−3.525	0.595	−0.287	0.011
$\Delta u/\text{m}\cdot\text{s}^{-1}$	298.15	−48.71	1.59	4.01	0.773
$\Delta k_S/\text{T}\cdot\text{Pa}^{-1}$	298.15	43.61	−11.59	−5.64	0.627
Ethyl Chloroacetate (1) + Anisole (2)					
$V^E \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	−0.416	−0.140	−0.025	0.005
	303.15	−0.035	0.271	−1.034	0.054
	308.15	−0.398	−0.109	0.165	0.005
$\Delta\eta/\text{mPa}\cdot\text{s}$	298.15	−0.149	−0.019	−0.013	0.003
	303.15	−0.139	−0.031	−0.001	0.002
	308.15	−0.118	−0.028	−0.040	0.001
$\Delta R \times 10^6/\text{m}^3\cdot\text{mol}^{-1}$	298.15	−0.256	0.020	0.156	0.006
	303.15	−0.115	−0.052	0.418	0.012
	308.15	−0.211	0.056	0.221	0.002
$\Delta u/\text{m}\cdot\text{s}^{-1}$	298.15	13.85	−0.56	−20.45	0.352
$\Delta k_S/\text{T}\cdot\text{Pa}^{-1}$	298.15	−27.01	−0.69	15.85	0.344

toluene > benzene > *p*-xylene > mesitylene. At higher temperatures, $\Delta\eta$ vs x_1 plots exhibit the same dependencies, but their magnitudes increase systematically with increasing temperature. This is typically displayed in Figure 3 for an ethyl chlroacetate + benzene mixture. For other mixtures, a similar trend is observed.

From the plots of ΔR vs ϕ_1 (volume fraction of ethyl chloroacetate) at 298.15 K shown in Figure 4, we find that a large negative deviation is observed for ethyl chloroacetate + mesitylene, while for ethyl chloroacetate + toluene or anisole, these values are slightly negative and overlap one another. The ΔR curves for ethyl chloroacetate + benzene are identical to those observed for the former two mixtures but are slightly more negative. For mixtures of ethyl chloroacetate + *p*-xylene or mesitylene, the ΔR curves are quite negative and more so in the case of ethyl chloroacetate + mesitylene. The effect of temperature on ΔR is not considerable in all of the binary mixtures, and hence these plots are not displayed.

The results of Δu vs x_1 are shown in Figure 5. For mixtures of ethyl chloroacetate + mesitylene, a negative deviation is observed, whereas for ethyl chloroacetate + anisole, the values are positive. However, in the case of mixtures of ethyl chloroacetate + benzene, toluene, or *p*-xylene, the curves show negative deviations and are skewed at the lower composition of ethyl chloroacetate.

Figure 6 displays the dependence of Δk_s on ϕ_1. Here, ethyl chloroacetate + mesitylene exhibits a positive deviation, while for anisole, a negative deviation is observed. With the remaining mixtures, the curves are more or less sigmoidal.

All of the quantities (V^E, $\Delta\eta$, ΔR, Δu, and Δk_s) have been fitted to the Redlich−Kister[15] equation by the method of least squares using the Marquardt algorithm[16] to derive the binary coefficient, A_i, and the standard deviation, σ:

$$V^E(\Delta Y) = x_1 x_2 \sum_{j=1}^{k} A_j (x_2 - x_1)^{j-1} \tag{4}$$

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

$$\sigma = \left[\sum (Y^E_{\text{cal}} - Y^E_{\text{obs}})^2/(n - m)\right]^{1/2} \tag{5}$$

where n represents the number of measurements and m the number of coefficients. The estimated values of A_j and σ for V^E, $\Delta\eta$, ΔR, Δu, and Δk_s are summarized in Table 3. In all cases, the best fit was obtained by using only three fitting coefficients. In all of the figures, the points represent the data calculated from eq 1, while smooth curves are drawn from the best-fitted data calculated from eq 4.

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