Density, Viscosity, Refractive Index, and Speed of Sound for the Binary Mixtures of Ethyl Chloroacetate with *n*-Alkanes (C_6 to C_{12}) at (298.15, 303.15, and 308.15) K

Jyoti N. Nayak, Mrityunjaya I. Aralaguppi,* and Tejraj M. Aminabhavi

Department of Chemistry, Karnatak University, Dharwad 580 003, India

Experimental data on density, viscosity, and refractive index at (298.15, 303.15, and 308.15) K and the speed of sound at 298.15 K are presented for the binary mixtures of ethyl chloroacetate + hexane, heptane, octane, nonane, decane, or dodecane. From these data, excess molar volume, deviations in viscosity, molar refraction, and isentropic compressibility have been calculated. The computed quantities have been fitted to Redlich−Kister equation to derive the coefficients and estimate the standard error values.

Introduction

The present paper is a part of our continuing study on the accumulation of binary mixture property data containing *n*-alkanes (Aminabhavi and Bindu, 1994; Aminabhavi et al., 1994, 1996, 1997; Aralaguppi et al., 1991, 1999). In this paper, ethyl chloroacetate (ECA) + *n*-alkane (hexane, heptane, octane, nonane, decane, and dodecane) mixtures have been studied. The alkanes have relevance in petrochemical industries, while ECA, due to its insecticidal activity, is used as a solvent in organic synthesis (Buckingham, 1982). Therefore, their binary mixture properties will be useful in industrial sectors. A survey of the literature indicates that no physical property data on these mixtures have been studied earlier. This prompted us to undertake a study on the measurement of density, ρ, viscosity, η, refractive index, n_D, and speed of sound, u, at different temperatures. Using these data, excess molar volume, V^E, and deviations in viscosity, $\Delta\eta$, molar refraction, ΔR, and isentropic compressibility, Δk_S, have been calculated. These results were further fitted to the Redlich−Kister equation (Redlich and Kister, 1948) to derive the binary coefficients and estimate the standard errors.

Experimental Section

Materials. High-purity spectroscopic and HPLC grade samples of hexane and heptane were procured from Spectrochem Ltd. and s.d. fine Chemicals, Mumbai, India. Octane was of analytical reagent grade while nonane, decane, and dodecane were all laboratory reagent grade and were procured from s.d. fine Chemicals, Mumbai, India. Ethyl chloroacetate was purchased from BASCO, Mumbai, India. The mole percent purities of all the liquids as determined by GC (HP 6890) using a FID detector were >99 and are reported in Table 1. Density and refractive index data at 298.15 K for the pure liquids are compared with the literature values in Table 1.

Binary mixtures were prepared by mass in specially designed glass-stoppered bottles (Aminabhavi et al., 1994). The mass measurements accurate to ±0.01 mg were done on a digital electronic balance (Mettler, AE 240, Switzerland). A set of nine compositions was prepared for each system, and their physical properties were measured at the respective compositions starting from 0.1 to 0.9 mole fraction in steps of 0.1. The possible error in mole fraction was estimated to be less than 10^{-4} in all the cases.

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,b]	1.4211[a]	1.4215[a,b]
hexane	(>99.9)	654.7	654.8[d]	1.3729	1.3723[c]
heptane	(>99.7)	679.3	679.5[d]	1.3859	1.3851[c]
octane	(>99.7)	698.5	698.5[d]	1.3952	1.3951[c]
nonane	(>98.0)	714.1	713.8[c]	1.4039	1.4031[c]
decane	(>99.0)	726.0	726.4[c]	1.4098	1.4096[c]
dodecane	(>99.0)	744.9	745.2[c]	1.4199	1.4195[c]

[a] Measured and compared at 293.15 K. [b] *CRC Handbook*, 1982−83. [c] Riddick et al., 1986. [d] Marsh, 1994.

Methods. Densities of liquids and liquid mixtures were measured to an accuracy of ±0.0001 using a capillary pycnometer of about 10 cm^3 volume as per the experimental details given earlier (Aralaguppi et al.,1991; Aminabhavi et al., 1994).

Viscosities were measured using a Cannon Fenske viscometer (size 75, ASTM D 445 supplied by 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. Calibrations of the pycnometer and viscometer are the same as described earlier (Aminabhavi and Bindu, 1994; Aminabhavi et al., 1994).

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

Speed of sound values were measured by using a variable-path single-crystal interferometer (Mittal Enterprises, New Delhi, Model M-84). A crystal-controlled high-frequency generator was used to excite the transducer at a frequency of 1 MHz. The frequency was measured within

* To whom correspondence should be addressed. E-mail: aralaguppi@yahoo.com.

10.1021/je010020w CCC: $20.00
Published on Web 06/01/2001

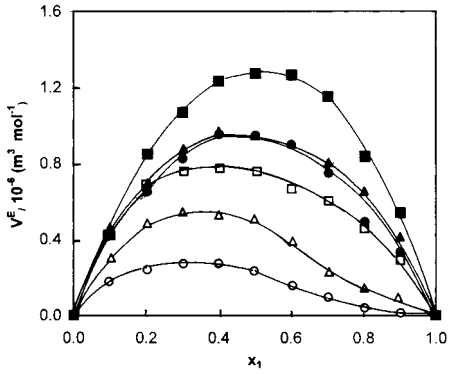

Figure 1. Excess molar volume (V^E) versus mole fraction of ethyl chloroacetate with (○) hexane, (△) heptane, (□) octane, (●) nonane, (▲) decane, and (■) dodecane at 298.15 K.

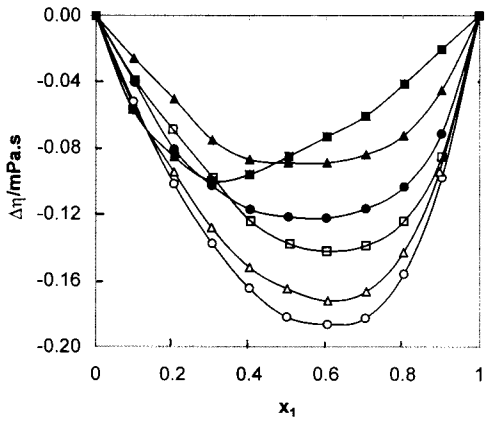

Figure 2. Deviations in viscosity ($\Delta\eta$) versus mole fraction at 298.15 K for mixtures of ethyl chloroacetate with alkanes. Symbols are the same as given in Figure 1.

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 thermostat maintained at 298.15 ± 0.01 K. To increase the accuracy of the measurement, several such maxima were counted by changing the distance between transducer and reflector. The total distance, d, moved by the reflector, which is fixed to the micrometer scale, corresponding to the two maxima in the ammeter, was measured and was used to calculate the wavelength, λ, by using $d = n\lambda/2$. By knowing the frequency, ν, of the crystal (1 MHz), speed of sound, u, in m·s^{-1} was calculated as $u = \nu\lambda$. The speed of sound values thus calculated are 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 the above measurements, the temperature was controlled within ±0.01 K using a constant temperature bath. A Julabo immersion cooler (FT 200, Julabo Labortechnik Gmbh, 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 into the bath liquid. At least three independent readings of the physical properties were taken for each composition, and the averages of these results are given in Table 2.

Results and Discussion

The results of V^E, $\Delta\eta$, ΔR, and Δk_S of the mixtures have been calculated respectively using the results of ρ, η, n_D, and u by using the relations (Aminabhavi and Bindu, 1994; Aminabhavi et al., 1996)

$$V^E = V_m - V_1x_1 - V_2x_2 \quad (1)$$

$$\Delta Y = Y_m - Y_1x_1 - Y_2x_2 \quad (2)$$

Here, V_m is molar volume of the mixture, 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, and ΔY represents $\Delta\eta$, ΔR, and Δk_S , respectively. Y_m is the

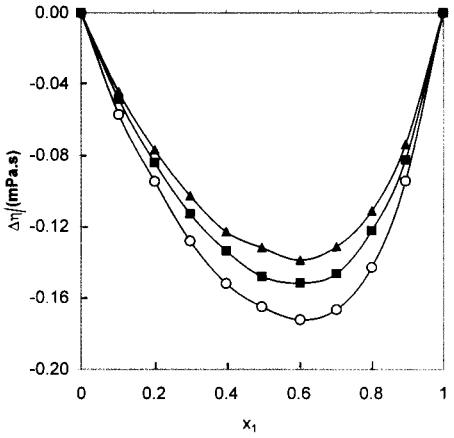

Figure 3. Effect of temperature on $\Delta\eta$ for the ethyl chloroacetate + heptane mixture: (○) 298.15 K; (■) 303.15 K; (▲) 308.15 K.

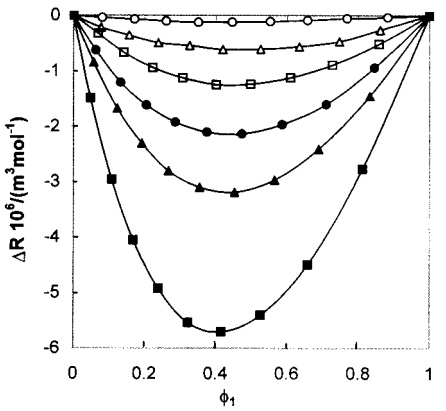

Figure 4. Deviations in molar refraction (ΔR) versus volume fraction at 298.15 K for mixtures of ethyl chloroacetate with alkanes. Symbols are the same as given in Figure 1.

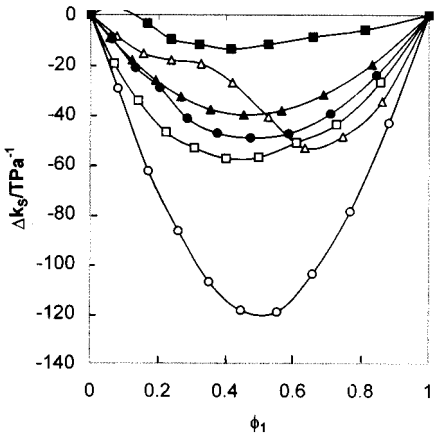

Figure 5. Deviations in isentropic compressibility (Δk_S) versus volume fraction at 298.15 K for mixtures of ethyl chloroacetate with alkanes. Symbols are the same as given in Figure 1.

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·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})
Ethyl Chloroacetate (1) + Hexane (2)									
298.15 K									
0.0000	654.7	0.318	1.3729	1081	0.6039	925.4	0.602	1.3988	1182
0.0977	693.5	0.342	1.3762	1089	0.7027	977.0	0.683	1.4040	1200
0.2029	737.5	0.375	1.3804	1102	0.8032	1031.9	0.787	1.4095	1218
0.3028	781.2	0.416	1.3844	1118	0.9022	1088.1	0.923	1.4147	1234
0.4001	825.6	0.465	1.3886	1137	1.0000	1146.1	1.097	1.4200	1249
0.4982	872.4	0.524	1.3933	1159					
303.15 K									
0.0000	649.4	0.303	1.3695		0.6039	919.4	0.566	1.3964	
0.0977	688.5	0.326	1.3733		0.7027	970.8	0.641	1.4016	
0.2029	732.2	0.357	1.3777		0.8032	1025.4	0.739	1.4070	
0.3028	775.8	0.396	1.3819		0.9022	1081.6	0.861	1.4123	
0.4001	820.0	0.441	1.3862		1.0000	1139.3	1.007	1.4173	
0.4982	866.7	0.495	1.3910						
308.15 K									
0.0000	645.0	0.288	1.3668		0.6039	913.4	0.530	1.3940	
0.0977	683.5	0.309	1.3706		0.7027	964.6	0.600	1.3992	
0.2029	726.9	0.339	1.3749		0.8032	1019.1	0.690	1.4046	
0.3028	770.3	0.377	1.3794		0.9022	1075.1	0.800	1.4098	
0.4001	814.4	0.417	1.3839		1.0000	1132.8	0.933	1.4147	
0.4982	860.9	0.466	1.3887						
Ethyl Chloroacetate (1) + Heptane (2)									
298.15 K									
0.0000	679.3	0.404	1.3859	1123	0.6048	921.8	0.651	1.4022	1160
0.1037	713.9	0.419	1.3873	1123	0.7050	973.4	0.726	1.4062	1188
0.2030	749.4	0.450	1.3897	1124	0.8022	1026.3	0.817	1.4100	1211
0.3009	787.1	0.484	1.3920	1125	0.8954	1080.4	0.930	1.4148	1233
0.4005	828.2	0.530	1.3955	1128	1.0000	1146.1	1.097	1.4200	1249
0.4992	871.6	0.585	1.3982	1139					
303.15 K									
0.0000	674.8	0.381	1.3827		0.6048	915.5	0.608	1.3995	
0.1037	709.3	0.398	1.3845		0.7050	967.0	0.676	1.4033	
0.2030	744.2	0.424	1.3867		0.8022	1020.0	0.761	1.4078	
0.3009	781.7	0.457	1.3893		0.8954	1073.6	0.859	1.4122	
0.4005	823.1	0.498	1.3923		1.0000	1139.3	1.007	1.4173	
0.4992	866.6	0.545	1.3955						
308.15 K									
0.0000	670.5	0.363	1.3801		0.6048	910.1	0.569	1.3966	
0.1037	704.7	0.378	1.3816		0.7050	961.1	0.634	1.4012	
0.2030	739.5	0.402	1.3841		0.8022	1014.0	0.710	1.4052	
0.3009	776.9	0.432	1.3863		0.8954	1067.6	0.800	1.4096	
0.4005	817.6	0.469	1.3898		1.0000	1132.8	0.933	1.4147	
0.4992	860.9	0.522	1.3928						
Ethyl Chloroacetate (1) + Octane (2)									
298.15 K									
0.0000	698.5	0.503	1.3952	1167	0.6021	916.4	0.718	1.4057	1209
0.1027	727.7	0.524	1.3966	1176	0.7042	966.3	0.782	1.4086	1217
0.2003	758.1	0.553	1.3974	1182	0.8031	1020.1	0.855	1.4119	1230
0.3048	794.1	0.586	1.3991	1189	0.9017	1079.3	0.953	1.4157	1241
0.4033	831.1	0.618	1.4010	1194	1.0000	1146.1	1.097	1.4200	1249
0.5064	873.3	0.666	1.4030	1202					
303.15 K									
0.0000	694.2	0.472	1.3932		0.6021	911.0	0.669	1.4032	
0.1027	723.1	0.493	1.3936		0.7042	961.2	0.723	1.4063	
0.2003	734.3	0.518	1.3948		0.8031	1013.7	0.796	1.4090	
0.3048	789.4	0.551	1.3961		0.9017	1072.9	0.884	1.4130	
0.4033	826.0	0.577	1.3982		1.0000	1139.3	1.007	1.4173	
0.5064	868.5	0.621	1.4006						
308.15 K									
0.0000	690.0	0.445	1.3906		0.6021	905.4	0.624	1.4003	
0.1027	718.8	0.465	1.3911		0.7042	955.2	0.676	1.4036	
0.2003	748.9	0.487	1.3922		0.8031	1008.5	0.741	1.4062	
0.3048	784.4	0.519	1.3938		0.9017	1066.9	0.820	1.4113	
0.4033	820.8	0.541	1.3955		1.0000	1132.8	0.934	1.4147	
0.5064	862.7	0.584	1.3981						
Ethyl Chloroacetate (1) + Nonane (2)									
298.15 K									
0.0000	714.1	0.656	1.4039	1211	0.6012	912.4	0.799	1.4088	1235
0.1004	739.2	0.660	1.4040	1212	0.7037	961.6	0.849	1.4109	1241
0.2033	768.1	0.665	1.4042	1215	0.8037	1016.3	0.907	1.4133	1245
0.3013	798.3	0.686	1.4050	1218	0.9012	1075.8	0.982	1.4164	1248
0.4018	832.3	0.716	1.4060	1225	1.0000	1146.1	1.097	1.4200	1249
0.5019	870.3	0.755	1.4073	1230					

Table 2. (Continued)

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})
				Ethyl Chloroacetate (1) + Nonane (2)					
				303.15 K					
0.0000	710.3	0.613	1.4013		0.6012	907.3	0.743	1.4060	
0.1004	734.7	0.620	1.4013		0.7037	956.8	0.792	1.4083	
0.2033	763.3	0.623	1.4017		0.8037	1010.0	0.842	1.4106	
0.3013	793.0	0.644	1.4024		0.9012	1069.3	0.912	1.4137	
0.4018	827.3	0.670	1.4033		1.0000	1139.3	1.007	1.4173	
0.5019	865.3	0.705	1.4048						
				308.15 K					
0.0000	706.2	0.575	1.3984		0.6012	902.1	0.688	1.4032	
0.1004	730.2	0.580	1.3987		0.7037	950.8	0.735	1.4056	
0.2033	758.6	0.583	1.3992		0.8037	1003.8	0.778	1.4078	
0.3013	788.2	0.602	1.4000		0.9012	1062.9	0.843	1.4112	
0.4018	822.3	0.624	1.4007		1.0000	1132.8	0.934	1.4147	
0.5019	860.2	0.654	1.4023						
				Ethyl Chloroacetate (1) + Decane (2)					
				298.15 K					
0.0000	726.0	0.820	1.4098	1228	0.6027	910.5	0.897	1.4118	1241
0.0992	748.0	0.821	1.4099	1229	0.7039	957.4	0.930	1.4130	1244
0.2037	774.4	0.825	1.4100	1231	0.8019	1009.9	0.969	1.4148	1246
0.3031	802.4	0.828	1.4103	1233	0.9008	1071.7	1.024	1.4170	1248
0.4005	833.2	0.843	1.4104	1236	1.0000	1146.1	1.097	1.4200	1249
0.5016	869.5	0.869	1.4112	1238					
				303.15 K					
0.0000	729.1	0.755	1.4071		0.6027	904.3	0.827	1.4092	
0.0992	743.8	0.761	1.4072		0.7039	951.6	0.854	1.4106	
0.2037	770.0	0.766	1.4074		0.8019	1003.9	0.885	1.4121	
0.3031	796.8	0.767	1.4075		0.9008	1065.9	0.946	1.4142	
0.4005	828.6	0.780	1.4077		1.0000	1139.3	1.007	1.4173	
0.5016	864.4	0.801	1.4086						
				308.15 K					
0.0000	717.0	0.711	1.4045		0.6027	899.5	0.765	1.4068	
0.0992	739.9	0.707	1.4047		0.7039	946.0	0.791	1.4078	
0.2037	765.4	0.713	1.4048		0.8019	998.3	0.829	1.4096	
0.3031	792.8	0.719	1.4048		0.9008	1059.6	0.872	1.4114	
0.4005	823.5	0.725	1.4054		1.0000	1132.8	0.934	1.4147	
0.5016	859.1	0.742	1.4060						
				Ethyl Chloroacetate (1) + Dodecane (2)					
				298.15 K					
0.0000	744.9	1.330	1.4199	1287	0.6026	904.0	1.116	1.4170	1264
0.0961	762.4	1.250	1.4191	1279	0.7029	948.0	1.105	1.4170	1258
0.2041	784.6	1.197	1.4182	1274	0.8040	1002.3	1.101	1.4172	1252
0.3012	807.8	1.159	1.4178	1272	0.9017	1065.5	1.099	1.4182	1250
0.4005	834.7	1.140	1.4175	1271	1.0000	1146.1	1.097	1.4200	1249
0.5029	867.2	1.127	1.4170	1267					
				303.15 K					
0.0000	740.9	1.209	1.4179		0.6026	899.5	1.029	1.4145	
0.0961	758.4	1.149	1.4169		0.7029	942.3	1.015	1.4145	
0.2041	780.8	1.094	1.4162		0.8040	996.2	0.994	1.4147	
0.3012	802.9	1.062	1.4156		0.9017	1058.8	0.985	1.4156	
0.4005	830.6	1.041	1.4150		1.0000	1139.3	1.007	1.4173	
0.5029	862.6	1.027	1.4147						
				308.15 K					
0.0000	737.6	1.111	1.4152		0.6026	893.9	0.940	1.4120	
0.0961	754.6	1.054	1.4148		0.7029	937.4	0.930	1.4120	
0.2041	776.3	1.019	1.4136		0.8040	989.9	0.927	1.4120	
0.3012	798.8	0.979	1.4131		0.9017	1053.3	0.929	1.4126	
0.4005	825.7	0.960	1.4128		1.0000	1132.8	0.934	1.4147	
0.5029	857.7	0.946	1.4121						

respective mixture property, viz., molar refractivity, R (calculated from the Lorenz−Lorentz relation), viscosity, η, speed of sound, u, and isentropic compressibility, k_S, for the binary mixtures; Y_i refers to pure component properties. While calculating the ΔR and Δk_S values, the volume fraction, ϕ_i, was used (Aminabhavi and Bindu, 1994; Aminabhavi et al., 1996, 1997). However, for the calculation of V^E and $\Delta\eta$, mole fraction was used.

The excess molar volume data displayed at 298.15 K in Figure 1 are positive in all the cases, indicating volume expansion upon mixing the alkanes with ethyl chloroacetate. The points on the curves represent the V^E values calculated from eq 1, while the smooth curves are drawn from the best fitted values of V^E calculated from eq 3. A large positive V^E (1.3 cm^3/mol) for the ethyl chloroacetate + dodecane mixture and a small V^E value of 0.23 cm^3/mol for the ethyl chloroacetate + hexane mixture signify that, with increasing size of alkane, the volume expansion becomes considerably larger. The equimolar values of V^E for mixtures of ethyl chloroacetate + nonane or + decane are almost identical, signifying not much difference in the interactions between these mixtures. The V^E results at other temperatures follow the same trends, but these are not displayed in order to avoid the redundancy of graphical presentations. However, there is no effect of temperature on excess volume.

The $\Delta\eta$ versus x_1 plots at 298.15 K displayed in Figure 2 show negative deviations in all the mixtures; also, these

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

function	temp/K	A_0	A_1	A_2	σ
Ethyl Chloroacetate (1) + Hexane (2)					
$10^6V^E/(m^3\cdot mol^{-1})$	298.15	0.893	1.071	0.110	0.013
	303.15	0.650	0.786	−0.260	0.015
	308.15	0.828	1.021	0.312	0.005
$\Delta\eta$/(mPa·s)	298.15	−0.727	0.288	−0.209	0.002
	303.15	−0.634	0.218	−0.114	0.002
	308.15	−0.573	0.204	−0.100	0.001
	298.15	−0.471	0.162	0.108	0.008
$10^6\Delta R/(m^3\cdot mol^{-1})$	303.15	−0.352	0.113	0.190	0.005
	308.15	−0.224	0.085	0.151	0.003
$\Delta k_S/(T\ Pa^{-1})$	298.15	−479.9	4.03	114.9	1.138
Ethyl Chloroacetate (1) + Heptane (2)					
$10^6V^E/(m^3\cdot mol^{-1})$	298.15	1.926	1.661	0.148	0.035
	303.15	1.946	1.676	0.675	0.051
	308.15	2.138	1.788	0.343	0.033
$\Delta\eta$/(mPa·s)	298.15	−0.665	0.246	−0.229	0.002
	303.15	−0.590	0.210	−0.168	0.002
	308.15	−0.526	0.184	−0.181	0.004
$10^6\Delta R/(m^3\cdot mol^{-1})$	298.15	−2.450	0.255	−0.216	0.018
	303.15	−2.400	0.299	0.170	0.014
	308.15	−2.371	0.367	−0.058	0.024
$\Delta k_S/(T\ Pa^{-1})$	298.15	−150.4	−162.6	−116.0	6.381
Ethyl Chloroacetate (1) + Octane (2)					
$10^6V^E/(m^3\cdot mol^{-1})$	298.15	2.984	1.008	1.562	0.020
	303.15	2.550	9.364	17.71	1.483
	308.15	3.118	1.307	1.038	0.018
$\Delta\eta$/(mPa·s)	298.15	−0.542	0.280	−0.198	0.006
	303.15	−0.482	0.244	−0.126	0.005
	308.15	−0.435	0.224	−0.128	0.006
$10^6\Delta R/(m^3\cdot mol^{-1})$	298.15	−4.966	0.912	0.341	0.010
	303.15	−5.392	−0.137	4.095	0.313
	308.15	−5.074	1.140	0.062	0.048
$\Delta k_S/(T\ Pa^{-1})$	298.15	−227.0	46.04	−52.12	0.791
Ethyl Chloroacetate (1) + Nonane (2)					
$10^6V^E/(m^3\cdot mol^{-1})$	298.15	3.792	0.592	−0.025	0.050
	303.15	3.881	1.461	1.155	0.073
	308.15	4.021	1.562	2.124	0.059
$\Delta\eta$/(mPa·s)	298.15	−0.489	0.129	−0.227	0.006
	303.15	−0.426	0.098	−0.137	0.003
	308.15	−0.401	0.111	−0.144	0.003
$10^6\Delta R/(m^3\cdot mol^{-1})$	298.15	−8.446	2.119	−0.633	0.020
	303.15	−8.465	1.961	−0.102	0.017
	308.15	−8.434	1.804	0.382	0.016
$\Delta k_S/(T\ Pa^{-1})$	298.15	−200.2	5.110	35.55	1.495
Ethyl Chloroacetate (1) + Decane (2)					
$10^6V^E/(m^3\cdot mol^{-1})$	298.15	3.805	0.3130	1.416	0.032
	303.15	7.687	7.003	9.738	0.218
	308.15	3.883	−0.362	−0.691	0.102
$\Delta\eta$/(mPa·s)	298.15	−0.364	0.096	−0.063	0.005
	303.15	−0.327	0.120	−0.017	0.007
	308.15	−0.320	0.095	−0.035	0.003
$10^6\Delta R/(m^3\cdot mol^{-1})$	298.15	−12.53	3.549	−0.442	0.001
	303.15	−11.44	1.697	2.299	0.111
	308.15	−12.49	3.520	−1.202	0.025
$\Delta k_S/(TPa^{1-})$	298.15	−159.1	21.60	9.430	0.200
Ethyl Chloroacetate (1) + Dodecane (2)					
$10^6V^E/(m^3\cdot mol^{-1})$	298.15	5.165	−0.386	0.448	0.041
	303.15	4.983	−0.692	1.112	0.067
	308.15	5.634	−0.244	1.181	0.036
$\Delta\eta$/(mPa·s)	298.15	−0.350	−0.234	−0.151	0.003
	303.15	−0.300	−0.132	−0.246	0.010
	308.15	−0.303	−0.110	−0.051	0.004
$10^6\Delta R/(m^3\cdot mol^{-1})$	298.15	−22.04	8.203	−3.501	0.037
	303.15	−22.13	8.429	−3.371	0.037
	308.15	−21.95	7.944	−3.131	0.028
$\Delta k_S/(T\ Pa^{-1})$	298.15	−55.77	4.59	72.75	2.86

data show a systematic increase from hexane to dodecane. For the ethyl chloroacetate + dodecane mixture, the $\Delta\eta$ values show a slight decreasing trend in the lower mole fraction range. At higher temperatures, $\Delta\eta$ versus x_1 plots exhibit the same dependencies, but these values increase systematically with increasing temperature for all the mixtures. A typical graph displaying the effect of temperature on the $\Delta\eta$ versus x_1 plot for the ethyl chloroacetate + heptane mixture is shown in Figure 3.

The results of ΔR and Δk_S versus ϕ_1 (volume fraction of ethyl chloroacetate) at 298.15 K are presented respectively in Figures 4 and 5. Both these parameters exhibit negative deviations in all the mixtures, further supporting weak (dispersion-type) intermolecular interactions between the mixing components. The values of ΔR decrease with increasing size of alkanes, while a reverse trend is seen for the dependence of Δk_S on volume fraction. The effect of temperature on ΔR is not considerable in all the binary mixtures, except ethyl chloroacetate + hexane mixtures, for which ΔR increases systematically with increasing temperature.

The mixing quantities, viz., V^E, $\Delta\eta$, ΔR, and Δk_S, have been fitted to the Redlich−Kister (Redlich and Kister, 1948) equation by the method of least-squares using the Marquardt algorithm (Marquardt, 1963) to derive the binary coefficient, A_i, and the standard deviation, σ:

$$V^E(\Delta Y) = x_1x_2\sum_{j=1}^{k}A_j(x_2 - x_1)^{j-1} \tag{3}$$

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,

$$\sigma = \left(\frac{\sum(Y^E_{cal} - Y^E_{obs})^2}{(n - m)}\right)^{1/2} \tag{4}$$

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, and Δk_S are summarized in Table 3. In all the cases, the best fit was obtained by using only three fitting coefficients in all the cases.

Acknowledgment

This research was funded by the Department of Science and Technology, New Delhi, India (SP/S1/H-09/200).

Literature Cited

Aminabhavi, T. M.; Bindu, G. Densities, Shear Viscosities, Refractive Indices, and Speeds of Sound of Bis(2-methoxyethyl) Ether with Nonane, Decane, Dodecane, Tetradecane, and Hexadecane at 298.15, 308.15, and 318.15 K. *J. Chem. Eng. Data* **1994**, *39*, 529−534.

Aminabhavi, T. M.; Aralaguppi, M. I.; Bindu, G.; Khinnavar, R. S. Densities, Shear Viscosities, Refractive Indices, and Speeds of Sound of Bis(2-methoxyethyl) Ether with Hexane, Heptane, Octane, and 2,2,4-Trimethylpentane in the Temperature Interval 298.15 to 318.15 K. *J. Chem. Eng. Data* **1994**, *39*, 522−528.

Aminabhavi, T. M.; Phayde, H. T. S.; Khinnavar, R. S.; Bindu, G. Densities, Refractive Indices, Speeds of Sound, and Shear Viscosities of Diethylene Glycol Dimethyl Ether with Ethyl Acetate, Methyl Benzoate, Ethyl Benzoate, and Diethyl Succinate in the Temperature Range from 298.15 to 318.15 K. *J. Chem. Eng. Data* **1994**, *39*, 251−260.

Aminabhavi, T. M.; Patil, V. B.; Aralaguppi, M. I.; Phayde, H. T. S. Density, Viscosity, and Refractive Index of the Binary Mixtures of Cyclohexane with Hexane, Heptane, Octane, Nonane, and Decane at (298.15, 303.15 and 308.15) K. *J. Chem. Eng. Data* **1996**, *41*, 521−525.

Aminabhavi, T. M.; Patil, V. B.; Aralaguppi, M. I.; Ortego, J. D.; Hansen, K. C. Density, Refractive Index, Viscosity, and Speed of Sound in Binary Mixtures of Ethenylbenzene with Hexane, Heptane, Octane, Nonane Decane, and Dodecane. *J. Chem. Eng. Data* **1997**, *42*, 641−646.

Aralaguppi, M. I.; Aminabhvi, T. M.; Balundgi, R. H.; Joshi, S. S. Thermodynamic Interactions in Mixtures of Bromoform with Hydrocarbons. *J. Phys. Chem.* **1991**, *95*, 5299−5308.

Aralaguppi, M. I.; Jadar, C. V.; Aminabhavi, T. M. Density, Refractive Index, Viscosity, and Speed of Sound in Binary Mixtures of 2-Ethoxyethanol with Dioxane, Acetonitrile, and Tetrahydrofuran, at (298.15, 303.15 and 308.15) K. *J. Chem. Eng. Data* **1996**, *41*, 1307−1310.

Aralaguppi, M. I.; Jadar, C. V.; Aminabhavi, T. M. Density, Viscosity, Refractive Index, and Speed of Sound in Binary Mixtures of 2-Chloroethanol with Methyl Acetate, Ethyl Acetate, *n*-Propyl Acetate, and *n*-Butyl Acetate. *J. Chem. Eng. Data* **1999**, *44*, 441−445.

Aralaguppi, M. I.; Jadar, C. V.; Aminabhavi, T. M. Density, Viscosity, Refractive Index, and Speed of Sound in Binary Mixtures of Cyclohexanone with Hexane, Heptane, Octane, Nonane, Decane, Dodecane, and 2,2,4-Trimethylpentane. *J. Chem. Eng. Data* **1999**, *44*, 435−440.

Buckingham, J., Ed. *Dictionary of Organic Compounds*, 3rd ed.; Chapman & Hall: London, 1982; Vol. 3.

CRC Handbook of Chemistry and Physics, 63rd ed.; Weast, R. C., Astle, M. J., Eds.; CRC Press: Boca Raton, FL, 1982−1983.

Marquardt, D. W. An Algorithm for Least Squares Estimation of Nonlinear Parameters. *J. Soc. Ind. Appl. Math.* **1963**, *11*, 431−441.

Marsh, K. N. *Data Bases for Chemistry and Engineering, TRC Thermodynamic Tables*; Texas A & M University System: 1994.

Redlich, O.; Kister, A. T. Algebraic Representation of Thermodynamic Properties and the Classification of Solutions. *Ind. Eng. Chem.* **1948**, *40*, 345−348.

Riddick, J. A.; Bunger, W. B.; Sakano, T. K. *Techniques of Chemistry, Organic Solvents. Physical Properties and Methods of Purifications*; John Wiley & Sons: New York, 1986; Vol. II.

Received for review January 17, 2001. Accepted April 5, 2001.

JE010020W