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

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Experimental values of density, viscosity, and refractive index at 298.15, 303.15, and 308.15 K and the speed of sound at 298.15 K in the binary mixtures of 2-chloroethanol with methyl acetate, ethyl acetate, propyl acetate, and butyl acetate are presented over the whole range of mixture composition. From these data, excess molar volume, deviations in viscosity, speed of sound, isentropic compressibility, and Lorenz−Lorentz molar refraction have been calculated. These results are fit to a Redlich−Kister type polynomial equation of the third degree to derive the binary coefficients. The values of standard deviations are estimated for the calculated and experimental data.

Introduction

2-Chloroethanol is a versatile solvent used in many industrial areas. It has the characteristics of both alcohol and chlorinated hydrocarbon and is soluble in water in all proportions and dissolves in many classes of compounds from inorganic salts to cellulose esters. Vapors of 2-chloroethanol possess highly toxic properties due probably to its hydrolysis in the tissue to form hydrochloric acid, and therefore, it should be handled only under extreme precautions for the protection against skin and breathing. Esters, on the other hand, find applications as plasticizers in polymer-processing industries in order to impart favorable thermoplastic behavior.

An understanding of the mixing behavior of 2-chloroethanol with esters is therefore important and has applications in many engineering areas. In an earliar study by Aminabhavi and Banerjee (1998; 1998a), the physicochemical property data are presented on the mixtures of 2-chloroethanol with *n*-alkanols. To the best of our knowledge, no extensive studies have been made on the mixtures of 2-chloroethanol with esters. In continuation of our ongoing program of research, we now report the results of density ρ, viscosity η, refractive index n_D with the sodium D-line, and speed of sound u for the binary mixtures of 2-chloroethanol with methyl acetate, ethyl acetate, propyl acetate, and butyl acetate over the entire range of mixture composition at 298.15, 303.15, and 308.15 K. The values of u are measured only at 298.15 K. By use of these data, the excess molar volume V^E, deviations in viscosity $\Delta\eta$, speed of sound Δu, isentropic compressibility Δk_S, and Lorenz−Lorentz molar refractivity ΔR have been computed. These data are graphically displayed in order to compare them with those calculated from the fittings with the Redlich and Kister (1948) polynomial equation. The standard deviations have been calculated between the experimental data and the calculated values for the investigated range of temperatures.

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

	ρ/g^{-1} cm^{-3}		n_D	
liquids (mol % purity)	expt	lit.	expt	lit.
2-chloroethanol (99.4)	1.1976	1.1965[a]	1.4402	1.4418[a]
methyl acetate (99.2)	0.9285	0.9282[b]	1.3602	1.3606[b]
ethyl acetate (99.4)	0.8948	0.8946[c]	1.3710	1.3714[d]
propyl acetate (99.3)	0.8831	0.8830[e]	1.3835	1.3828[e]
butyl acetate (99.5)	0.8757	0.8759[f]	1.3931	1.3929[f]

[a] Riddick et al. (1986). [b] Aminabhavi and Banerjee (1998a). [c] Ortega et al. (1987). [d] Aminabhavi et al. (1994). [e] Correa et al. (1991). [f] Aminabhavi et al. (1993).

Experimental Section

Materials. High-purity spectroscopic and analytical grade samples of methyl acetate and ethyl acetate were procured from s.d. Fine Chemicals Ltd., Mumbai, India. 2-Chloroethanol, propyl acetate, and butyl acetate were purchased from E. Merck (Germany). All the samples were used without further purification because their purities exceeded 99% as tested by gas chromatography (HP 6890 series) using a flame ionization detector with a packed column (see Table 1). Experimental values of ρ and n_D along with the mol % purity of the liquids at 298.15 K are given in Table 1. These results agreed well with the published data. Mixtures were prepared by mass in specially designed glass-stoppered bottles, and the measurements were performed on the same day. An electronic Mettler balance, model AE 240 with a precision of ±0.01 mg was used. The error in mole fraction is around ±0.0002.

Measurements. Experimental details about the preparation of binary mixtures, measurements of mass, density, refractive index, speed of sound, and viscosity of pure liquids and binary mixtures are the same as described previously (Aralaguppi et al., 1991). The binary mixtures were prepared by mass by mixing the calculated volumes of liquid components in airtight glass bottles. The mass measurements (±0.01 mg) were made using an electronic balance (Mettler AE 240, Switzerland). A set of nine compositions were prepared for each system, and their

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

x_1	ρ/(g^{-1} cm^{-3})	n_D	η/(mPa s)	u/(m s^{-1})	x_1	ρ/(g^{-1} cm^{-3})	n_D	η/(mPa s)	u/(m s^{-1})
				2-Chloroethanol (1) + Methyl Acetate (2)					
				298.15 K					
0.0000	0.9285	1.3602	0.384	1159	0.6047	1.0806	1.4081	1.039	1256
0.1021	0.9521	1.3682	0.438	1174	0.7046	1.1087	1.4162	1.324	1276
0.2013	0.9759	1.3759	0.501	1187	0.7987	1.1360	1.4240	1.688	1296
0.3049	1.0015	1.3840	0.587	1203	0.9014	1.1668	1.4325	2.234	1324
0.4029	1.0266	1.3918	0.691	1220	1.0000	1.1976	1.4402	2.985	1363
0.5056	1.0537	1.4000	0.844	1238					
				303.15 K					
0.0000	0.9218	1.3580	0.365		0.6047	1.0743	1.4072	0.954	
0.1021	0.9457	1.3669	0.414		0.7046	1.1027	1.4139	1.197	
0.2013	0.9696	1.3734	0.472		0.7987	1.1306	1.4223	1.518	
0.3049	0.9954	1.3812	0.555		0.9014	1.1612	1.4305	1.978	
0.4029	1.0204	1.3893	0.645		1.0000	1.1921	1.4373	2.580	
0.5056	1.0478	1.3979	0.787						
				308.15 K					
0.0000	0.9152	1.3547	0.349		0.6047	1.0688	1.4038	0.881	
0.1021	0.9392	1.3632	0.392		0.7046	1.0971	1.4120	1.094	
0.2013	0.9633	1.3713	0.447		0.7987	1.1247	1.4197	1.369	
0.3049	0.9891	1.3796	0.519		0.9014	1.1558	1.4276	1.762	
0.4029	1.0143	1.3874	0.606		1.0000	1.1866	1.4342	2.261	
0.5056	1.0416	1.3957	0.726						
				2-Chloroethanol (1) + Ethyl Acetate (2)					
				298.15 K					
0.0000	0.8948	1.3710	0.430	1148	0.6007	1.0489	1.4080	1.065	1238
0.1033	0.9171	1.3771	0.485	1158	0.7022	1.0822	1.4152	1.326	1263
0.2006	0.9397	1.3822	0.542	1171	0.8000	1.1169	1.4232	1.689	1290
0.3231	0.9700	1.3894	0.643	1186	0.9000	1.1554	1.4320	2.242	1322
0.4048	0.9916	1.3948	0.729	1198	1.0000	1.1976	1.4402	2.985	1363
0.5015	1.0189	1.4013	0.864	1215					
				303.15 K					
0.0000	0.8887	1.3681	0.407		0.6007	1.0433	1.4059	0.967	
0.1033	0.9110	1.3742	0.454		0.7022	1.0764	1.4132	1.200	
0.2006	0.9336	1.3799	0.510		0.8000	1.1111	1.4217	1.517	
0.3231	0.9640	1.3874	0.601		0.9000	1.1498	1.4301	1.980	
0.4048	0.9858	1.3937	0.679		1.0000	1.1921	1.4373	2.580	
0.5015	1.0130	1.3988	0.800						
				308.15 K					
0.0000	0.8827	1.3654	0.387		0.6007	1.0374	1.4034	0.894	
0.1033	0.9051	1.3716	0.430		0.7022	1.0707	1.4110	1.098	
0.2006	0.9277	1.3775	0.482		0.8000	1.1056	1.4191	1.369	
0.3231	0.9581	1.3845	0.565		0.9000	1.1443	1.4270	1.776	
0.4048	0.9798	1.3897	0.635		1.0000	1.1866	1.4342	2.261	
0.5015	1.0072	1.3962	0.746						
				2-Chloroethanol (1) + Propyl Acetate (2)					
				298.15 K					
0.0000	0.8831	1.3835	0.551	1172	0.5974	1.0286	1.4106	1.155	1249
0.1017	0.9024	1.3872	0.593	1178	0.7006	1.0642	1.4176	1.430	1268
0.2031	0.9235	1.3912	0.658	1190	0.7963	1.1014	1.4246	1.805	1290
0.3017	0.9461	1.3955	0.736	1201	0.9006	1.1474	1.4326	2.302	1318
0.4061	0.9724	1.4003	0.852	1218	1.0000	1.1976	1.4402	2.985	1363
0.5057	1.0002	1.4058	0.989	1232					
				303.15 K					
0.0000	0.8774	1.3807	0.512		0.5974	1.0219	1.4081	1.057	
0.1017	0.8970	1.3846	0.557		0.7006	1.0587	1.4151	1.293	
0.2031	0.9180	1.3885	0.614		0.7963	1.0983	1.4222	1.615	
0.3017	0.9406	1.3931	0.687		0.9006	1.1420	1.4302	2.035	
0.4061	0.9668	1.3980	0.792		1.0000	1.1921	1.4373	2.580	
0.5057	0.9948	1.4036	0.908						
				308.15 K					
0.0000	0.8718	1.3783	0.483		0.5974	1.0176	1.4059	0.970	
0.1017	0.8912	1.3820	0.525		0.7006	1.0532	1.4129	1.177	
0.2031	0.9123	1.3861	0.580		0.7963	1.0903	1.4200	1.473	
0.3017	0.9349	1.3906	0.642		0.9006	1.1363	1.4279	1.810	
0.4061	0.9613	1.3956	0.733		1.0000	1.1866	1.4342	2.261	
0.5057	0.9892	1.4006	0.856						

Table 2 (Continued)

x_1	ρ/(g^{-1} cm^{-3})	n_D	η/(mPa s)	u/(m s^{-1})	x_1	ρ/(g^{-1} cm^{-3})	n_D	η/(mPa s)	u/(m s^{-1})
2-Chloroethanol (1) + Butyl Acetate (2)									
298.15 K									
0.0000	0.8757	1.3931	0.662	1190	0.5993	1.0137	1.4140	1.286	1250
0.1068	0.8940	1.3959	0.718	1198	0.7000	1.0493	1.4195	1.549	1269
0.2078	0.9132	1.3989	0.784	1209	0.8008	1.0910	1.4252	1.919	1289
0.3045	0.9336	1.4020	0.865	1218	0.8994	1.1390	1.4330	2.359	1321
0.4056	0.9577	1.4057	0.970	1228	1.0000	1.1976	1.4402	2.985	1363
0.5002	0.9833	1.4095	1.105	1238					
303.15 K									
0.0000	0.8706	1.3907	0.623		0.5993	1.0084	1.4118	1.169	
0.1068	0.8887	1.3934	0.669		0.7000	1.0442	1.4172	1.391	
0.2078	0.9077	1.3964	0.732		0.8008	1.0856	1.4232	1.704	
0.3045	0.9284	1.3996	0.801		0.8994	1.1337	1.4308	2.106	
0.4056	0.9525	1.4033	0.894		1.0000	1.1921	1.4373	2.580	
0.5002	0.9781	1.4071	1.013						
308.15 K									
0.0000	0.8655	1.3882	0.584		0.5993	1.0031	1.4093	1.070	
0.1068	0.8837	1.3910	0.627		0.7000	1.0388	1.4146	1.262	
0.2078	0.9028	1.3939	0.684		0.8008	1.0803	1.4209	1.518	
0.3045	0.9233	1.3967	0.746		0.8994	1.1282	1.4283	1.849	
0.4056	0.9473	1.4008	0.828		1.0000	1.1866	1.4342	2.261	
0.5002	0.9728	1.4047	0.932						

physical properties were measured the same day. The reproducibility in mole fraction was within ±0.0001 units. Densities of pure liquids and their mixtures were measured using a double-arm pycnometer having a bulb volume of 15 cm^3 and a capillary bore with an internal diameter of 1 mm. Doubly distilled, deionized, and degassed water with a specific conductance of 1×10^{-4} Ω^{-1} cm^{-1} was used for calibration. For all the mixtures and pure solvents, triplicate measurements were performed and the average of these values was considered in all calculations. Density values are accurate to ±0.0002 g cm^{-3}.

Viscosities were measured using a Cannon Fenske viscometer (size 75, ASTM D 445 supplied by Industrial Research Glassware Ltd., Roselle, New Jersey). 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, 1995; Aminabhavi et al., 1994).

Refractive indices for the sodium D-line were measured using a thermostatically controlled Abbe refractometer (Atago 3T, made in Japan). A minimum of three independent readings were taken for each composition, and their average is taken for calculation. The refractive index values are accurate to ±0.0001 units. The speed of sound values were measured using a variable-path single-crystal interferometer (model M-84, Mittal Enterprises, New Delhi). The interferometer was used at a frequency of 1 kHz and was calibrated using benzene and toluene. The speed of sound values are accurate to ±2 m s^{-1}. In all the property measurements, an INSREF, model 016 AP thermostat was used at a constant digital temperature display accurate to ±0.01 K. The results of ρ, n_D, η, and u compiled in Table 2 represent the averages of three independent measurements for each composition of the mixture.

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 in the bath liquid.

Results and Discussion

From the results of densities given in Table 2, excess molar volumes have been calculated as

$$V^E = V_m - V_1 x_1 - V_2 x_2 \quad (1)$$

where V_m is molar volume of the mixture, V_1 and V_2 are the molar volumes of components 1 and 2 of the mixture, and x_i represents the mole fraction of the ith component in the mixture. From the values of η, n_D, u, and k_S of the individual components as well as of the binary mixtures, the $\Delta\eta$, ΔR, Δu, and Δk_S have been calculated from

$$\Delta Y = Y_m - Y_1 c_1 - Y_2 c_2 \quad (2)$$

For calculating $\Delta\eta$ and Δu, we have used the mole fractions x_i for c_i. However, for calculating ΔR and Δk_S, the volume fraction

$$\phi_i = \frac{x_i V_i}{\sum_i x_i V_i}$$

was used for c_i.

Each set of excess functions have been fit to the Redlich and Kister (1948) polynomial

$$V^E \text{ (or } \Delta Y) = c_1 c_2 \sum_{j=1}^{k} A_j (c_2 - c_1)^{j-1} \quad (3)$$

to estimate the parameter values of A_0, A_1, and A_2 by the method of least squares using the Marquardt algorithm (1963). It was found that the best fittings were obtained for the solution of eq 3 with only three adjustable parameters.

The standard deviations σ between the fit quantities of eq 3 and the computed quantities eqs 1 and 2 were calculated as

$$\sigma = \left(\frac{\sum (V^E_{cal} \text{ (or } \Delta Y_{cal}) - V^E_{obs} \text{ (or } \Delta Y_{obs}))^2}{n - m} \right)^{1/2} \quad (4)$$

Table 3. Derived Parameters of Excess Functions for Mixtures

function	temp/K	A_0	A_1	A_2	σ
2-Chloroethanol (1) + Methyl Acetate (2)					
$V^E \times 10^6/(m^3\ mol^{-1})$	298.15	−0.158	0.011	0.097	0.001
	303.15	−0.237	−0.085	−0.048	0.014
	308.15	−0.326	−0.030	−0.037	0.002
$\Delta\eta$/(mPa s)	298.15	−3.401	1.920	−0.776	0.006
	303.15	−2.800	1.474	−0.504	0.004
	308.15	−2.350	1.156	−0.353	0.002
$\Delta R \times 10^6/(m^3\ mol^{-1})$	298.15	0.506	−0.001	0.153	0.002
	303.15	0.566	0.198	0.246	0.029
	308.15	0.713	0.052	0.309	0.001
$\Delta u/(m\ s^{-1})$	298.15	−97.75	66.36	−69.28	1.370
$\Delta k_S/(TPa^{-1})$	298.15	−72.54	47.56	37.50	0.773
2-Chloroethanol (1) + Ethyl Acetate (2)					
$V^E \times 10^6/(m^3\ mol^{-1})$	298.15	−0.250	−0.134	−0.020	0.003
	303.15	−0.333	−0.150	0.140	0.007
	308.15	−0.369	−0.129	0.031	0.003
$\Delta\eta$/(mPa s)	298.15	−3.376	1.958	−0.805	0.008
	303.15	−2.792	1.519	−0.528	0.004
	308.15	−2.335	1.185	−0.330	0.009
$\Delta R \times 10^6/(m^3\ mol^{-1})$	298.15	−1.400	0.281	0.169	0.013
	303.15	−1.261	0.304	0.461	0.021
	308.15	−1.250	0.428	0.452	0.009
$\Delta u/(m\ s^{-1})$	298.15	−158.1	46.46	−8.33	1.216
$\Delta k_S/(TPa^{-1})$	298.15	−92.43	2.095	−1.195	1.159
2-Chloroethanol (1) + Propyl Acetate (2)					
$V^E \times 10^6/(m^3\ mol^{-1})$	298.15	0.048	0.054	0.014	0.001
	303.15	0.109	0.213	−1.071	0.069
	308.15	−0.073	0.079	0.145	0.003
$\Delta\eta$/(mPa s)	298.15	−3.161	1.631	−0.561	0.010
	303.15	−2.585	1.252	−0.292	0.008
	308.15	−2.147	0.955	−0.144	0.014
$\Delta R \times 10^6/(m^3\ mol^{-1})$	298.15	−4.634	1.453	−0.138	0.007
	303.15	−4.575	1.371	−0.271	0.017
	308.15	−4.568	1.629	0.042	0.008
$\Delta u/(m\ s^{-1})$	298.15	−141.1	69.04	−99.22	1.493
$\Delta k_S/(TPa^{-1})$	298.15	−108.6	33.92	64.97	1.145
2-Chloroethanol (1) + Butyl Acetate (2)					
$V^E \times 10^6/(m^3\ mol^{-1})$	298.15	0.334	−0.019	−0.129	0.002
	303.15	0.281	0.121	0.077	0.007
	308.15	0.256	−0.005	−0.177	0.002
$\Delta\eta$/(mPa s)	298.15	−2.884	1.406	−0.356	0.007
	303.15	−2.382	1.037	−0.080	0.010
	308.15	−1.973	0.872	−0.153	0.004
$\Delta R \times 10^6/(m^3\ mol^{-1})$	298.15	−8.874	3.129	−0.895	0.014
	303.15	−8.846	3.256	−0.846	0.013
	308.15	−8.818	3.374	−0.917	0.017
$\Delta u/(m\ s^{-1})$	298.15	−151.9	116.0	−58.19	0.826
$\Delta k_S/(TPa^{-1})$	298.15	−84.41	49.40	−42.90	0.866

where n represents the number of data points (11) and m the number of coefficients (3). The fitted parameter values along with the standard deviations are presented in Table 3. It is to be noted that in Figures 1−5, the best solid lines of eq 3 are drawn from the smoothed computed values, whereas the points represent those calculated from eqs 1 and 2.

Excess molar volumes of 2-chloroethanol + esters at 298.15 K are displayed in Figure 1. The values of V^E are negative for the binary mixtures of 2-chloroethanol + methyl acetate, or + ethyl acetate. For the mixtures of 2-chloroethanol + propyl acetate, or + butyl acetate, the V^E values are positive. On the other hand, between mixtures of 2-chloroethanol with methyl acetate or ethyl acetate, the former exhibits a somewhat higher V^E than the latter. Similar dependencies are observed at higher temperatures (i.e., 303.15 and 308.15 K), but these plots are not presented in order to reduce the number of graphs.With increasing temperature, in the case of 2-chloroethanol + methyl acetate, or + ethyl acetate, the negative values of V^E become more negative. On the other hand, for mixtures of 2-chloroethanol + propyl acetate, or + butyl acetate, the positive V^E values become more positive at higher temperatures.

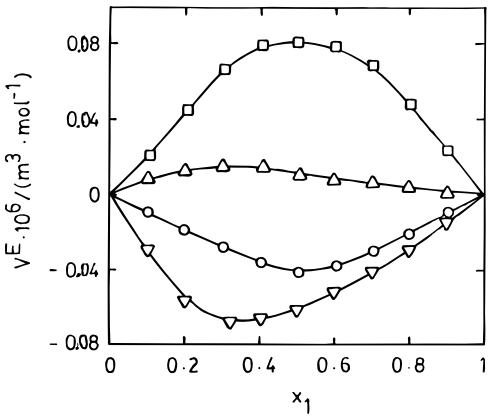

Figure 1. Plots of excess molar volume vs mole fraction of 2-chloroethanol at 298.15 K for the mixtures of 2-chloroethanol with (○) methyl acetate, (▽) ethyl acetate, (△) propyl acetate, (□) butyl acetate.

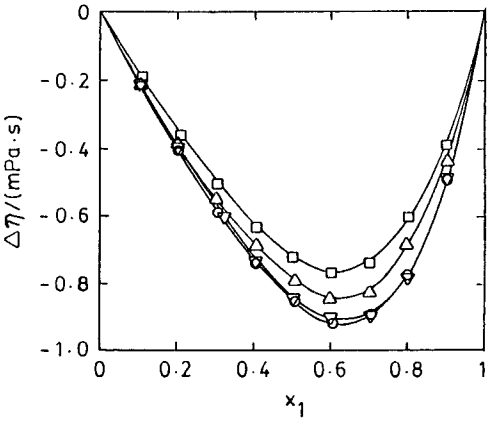

Figure 2. Plots of deviation in viscosity vs mole fraction of 2-chloroethanol at 298.15 K for the same mixtures given in Figure 1.

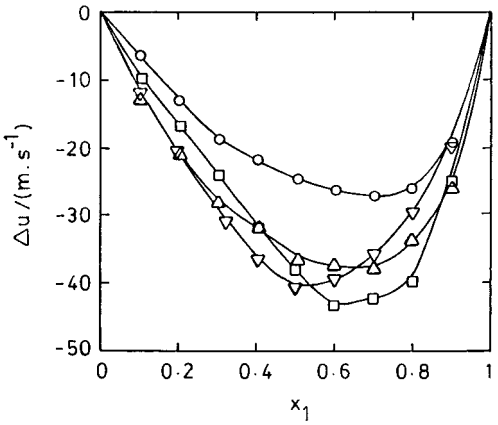

Figure 3. Plots of deviation in speed of sound vs mole fraction of 2-chloroethanol at 298.15 K for the same mixtures given in Figure 1.

The plots of $\Delta\eta$ vs x_1 for all the binary mixtures at 298.15 K are displayed in Figure 2. For all the mixtures, the $\Delta\eta$ values are negative. For 2-chloroethanol + methyl acetate, or + ethyl acetate mixtures, the negative $\Delta\eta$ values are almost identical over the entire range of mixture composition, and hence, their dependencies are shown by a single curve. The $\Delta\eta$ values for mixtures of 2-chloroethanol + propyl acetate are slightly more negative than those

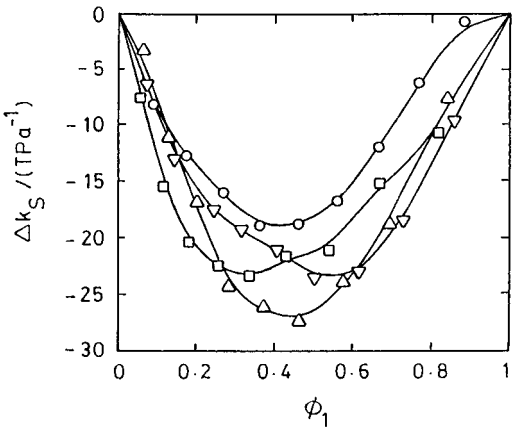

Figure 4. Plots of deviation in isentropic compressibility vs volume fraction of 2-chloroethanol at 298.15 K for the same mixtures as given in Figure 1.

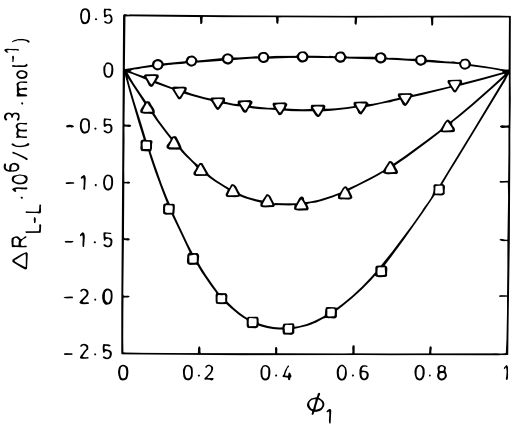

Figure 5. Plots of deviation in Lorenz−Lorentz molar refraction vs volume fraction of 2-chloroethanol at 298.15 K for the same mixtures given in Figure 1.

observed for the 2-chloroethanol + butyl acetate mixture. In all the mixtures, the $\Delta\eta$ values increase with increasing temperature, but the curves are not presented.

The results of Δu vs x_1 at 298.15 K are displayed in Figure 3 wherein we observe that the Δu values are negative in all the cases. Also, the curves do not show any systematic variation with the size of the esters.

The plots of Δk_S vs ϕ_1 at 298.15 K displayed in Figure 4 are also negative for all the mixtures. On the other hand, the results of ΔR vs ϕ_1 at 298.15 K presented in Figure 5 are positive for 2-chloroethanol + methyl acetate, while the negative ΔR values are observed for the remaining mixtures. Also, these values decrease systematically with increasing size of the molecules, i.e., from methyl acetate to butyl acetate. The ΔR vs ϕ_1 curves at 303.15 and 308.15 K show the same trends, but these are not presented. However, the effect of temperature on ΔR values is significant only in the case of mixtures of 2-chloroethanol + methyl acetate, but for higher esters, the effect is almost insignificant.

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