Relative Permittivities, Densities, Refractive Indices and Ultrasound Velocities of the Binary Systems of γ-Butyrolactone with Methanol, Ethanol, 1-Butanol, and 1-Octanol

George Ritzoulis,*,† Doukeni Missopolinou,‡ Stauroula Doulami,† and Costas Panayiotou*,‡

Departments of Chemistry and Chemical Engineering, Aristotle University of Thessaloniki, 54006 Thessaloniki, Greece

Relative permittivities, ϵ, densities, ρ, refractive indices, n_D, and ultrasonic velocities, u, were measured for the binary systems of γ-butyrolactone with methanol, ethanol, and 1-octanol at 298.15 K and with 1-butanol at 298.15 K and 303.15 K, over the entire composition range. From the experimental results, the excess volume, V^E, and the deviations in relative permittivity, $\Delta\epsilon$, ultrasonic velocity, Δu, isentropic compressibility, Δk_s, and molar polarization, ΔP_m, were calculated. The above functions show deviations from the additivity law, which depend on the length of the carbon chain of the alcohol.

Introduction

The aim of this work is to study some physicochemical properties of the binary systems of γ-butyrolactone (γ-BL) with the alkanols: methanol (MeOH); ethanol (EtOH); 1-butanol (BuOH); 1-octanol (OcOH).

γ-Butyrolactone is an important solvent having several advantageous physicochemical properties such as a broad liquid range (−45 °C to 205 °C) and medium relative permittivity (41.65 at 25 °C). It has a fairly high viscosity (1.76 mPa·s at 25 °C), and it is an excellent electrolytic solvent having wide applications as a medium for many chemical and electrochemical reactions. The usefulness of γ-butyrolactone as a solvent in lithium batteries is broadly known.[1,2] Mixtures of γ-butyrolactone with other solvents are also of particular interest. Research on their miscellaneous properties has been done in various laboratories.[3−9]

The measured quantities in this work are relative permittivity, ϵ, density, ρ, refractive index, n_D, and ultrasonic velocity, u, over the entire range of mole fractions at 298.15 K. For the system γ-butyrolactone + 1-butanol, in particular, the above quantities were studied at temperatures 298.15 K and 303.15 K, to examine the effect of temperature on the above properties. The measured experimental values of the ultrasonic velocity along with those of density are used to calculate the isentropic compressibility coefficient, k_s, which provides useful information about the interactions that take place between the components of the mixture. Also, from the experimental values of relative permittivity and refractive index, the molar polarization, P_m, has been estimated.

The excess volume, V^E, the deviations in the relative permittivity, $\Delta\epsilon$, ultrasonic velocity, Δu, isentropic compressibility, Δk_s, and polarity, ΔP_m, have also been calculated. The resulting functions are fitted to Redlich−Kister type polynomial equation, and the adjustable parameters along with the standard deviation are also given.

Materials and Methods

γ-Butyrolactone (Aldrich, >99 mol %) was purified by distillation under reduced pressure, as has been proposed by Aurbach and Gottlieb.[10] Methanol (Merck >99.8 mol %), ethanol (Merck 99.8 mol %), 1-butanol (Aldrich 99.8 mol %), and 1-octanol (Fluka 99.5 mol %) were used as received without further purification but were kept over molecular sieves 3 and 4 Å. The purities of the alkanols were checked by gas chromatography (Carlo Erba Vega Series 2) equipped with a capillary column (J&W, DB-1, 30 m × 0.32 mm × 0.25 μm) and flame ionization detector.

The binary mixtures were prepared by weight and degassed on ultrasonic bath. The accuracy of the mole fraction was estimated to be ±0.0002. Densities were measured with an Anton-Paar 60/602 vibrating tube densitometer, which was thermostated by a Haake F3-K digital thermostat, with a temperature stability better than ±0.01 K. The accuracy in density measurements was better than

Table 1. Experimental and Literature Values of Pure Liquids at 298.15 K

liquid	ρ (g·cm^{-3})	ϵ	n_D	u (m·s^{-1})
γ-butyrolactone	1.1239[a]	41.68[a]	1.4355[a]	1520.5[a]
	1.1237[b]	41.7[b]		1510[b]
		41.78[s]		
methanol	0.7866[a]	32.64[a]	1.3266[a]	1101.6[a]
	0.7866[d]	32.65[q]	1.3275[e]	1108[e]
	0.7868[e]		1.3274[f]	
	0.78655[g]		1.3265[n]	
	0.78637[h]			
	0.7866[i]			
ethanol	0.7853[a]	24.55[a]	1.3597[a]	1142.6[a]
	0.7862[e]	24.33[q]	1.3608[e]	1142.4[m]
	0.7861[k]		1.3595[f]	1148[p]
	0.78524[g]		1.3593[g]	
	0.7853[j]		1.3592[n]	
1-butanol	0.8060[a]	17.10[a]	1.3973[a]	1239.4[a]
	0.8071[e]	17.7[q]	1.3984[e]	1244[p]
	0.8056[k]		1.3974[f]	
	0.80577[g]		1.3973[o]	
	0.80585[l]			
1-octanol	0.8221[a]	10.02[a]	1.4284[a]	1347.4[a]
	0.8222[k]	9.85[q]	1.4275[r]	

[a] This work. [b] Ramkumar et al.[5] [c] Pistoia.[1] [d] Won et al.[12] [e] Aminabhavi et al.[13] [f] Ortega et al.[14] [g] Francesconi et al.[15] [h] Riddick et al.[16] [i] Martin et al.[17] [j] El-Banna et al.[18] [k] Rauf et al.[19] [l] Artigas et al.[20] [m] Papaioannou et al.[11] [n] Rodriguez et al.[21] [o] Marsh.[22] [p] Aralaguppi et al.[23] [q] Landolt−Börnstein.[24] [r] Arce et al.[25] [s] Côté et al.[6]

* Corresponding authors.
† Department of Chemistry.
‡ Department of Chemical Engineering.

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Table 2. Experimental Density, ρ, Relative Permittivity, ϵ, Refractive Index, n_D, and Ultrasonic Velocities, u, for the Binary Systems γ-BL + Alkanol

x_1	ρ (g·cm^{-3})	x_1	ϵ	x_1	n_D	x_1	u (m·s^{-1})
			(i) T = 298.15 K, γ-Butyrolactone (1) + Methanol (2)				
0.0000	0.7866	0.0000	32.64	0.0000	1.3266	0.0000	1101.6
0.1000	0.8495	0.1000	35.33	0.1000	1.3483	0.1000	1174.5
0.2011	0.9027	0.2011	36.71	0.2011	1.3689	0.2011	1238.3
0.3000	0.9460	0.3000	37.91	0.3000	1.3830	0.3000	1292.7
0.4002	0.9836	0.4002	38.95	0.4002	1.3952	0.4002	1339.1
0.5000	1.0160	0.5000	39.71	0.5000	1.4069	0.5000	1379.9
0.6005	1.0445	0.6005	40.36	0.6005	1.4150	0.6005	1416.1
0.7000	1.0690	0.7000	40.89	0.7000	1.4250	0.7000	1448.0
0.7999	1.0903	0.7999	41.24	0.7999	1.4273	0.7999	1475.1
0.9000	1.1081	0.9000	41.43	0.9000	1.4330	0.9000	1498.8
1.0000	1.1239	1.0000	41.68	1.0000	1.4355	1.0000	1520.5
			(ii) T = 298.15 K, γ-Butyrolactone (1) + Ethanol (2)				
0.0000	0.7853	0.0000	24.55	0.0000	1.3597	0.0000	1142.6
0.1000	0.8298	0.1000	26.52	0.1000	1.3707	0.1000	1193.0
0.2000	0.8714	0.2176	28.82	0.2176	1.3828	0.2000	1239.9
0.3000	0.9106	0.3001	30.36	0.3001	1.3901	0.3000	1282.4
0.4000	0.9477	0.4133	32.49	0.4133	1.3991	0.4000	1322.7
0.5000	0.9821	0.4991	34.11	0.4991	1.4065	0.5000	1361.9
0.6000	1.0143	0.6002	35.79	0.6002	1.4131	0.6000	1398.3
0.7001	1.0446	0.6984	37.38	0.6984	1.4196	0.7001	1433.8
0.7994	1.0727	0.7998	38.97	0.7998	1.4256	0.7994	1467.8
0.9000	1.0984	0.9000	40.38	0.9000	1.4310	0.8901	1493.9
1.0000	1.1239	1.0000	41.66	1.0000	1.4358	1.0000	1520.5
			(iii) T = 298.15 K, γ-Butyrolactone (1) + 1-Octanol (2)				
0.0000	0.8221	0.0000	10.02	0.0000	1.4284	0.0000	1347.4
0.1000	0.8379	0.1000	11.19	0.1000	1.4285	0.1000	1353.6
0.2003	0.8491	0.2035	12.23	0.2035	1.4288	0.2035	1357.8
0.3016	0.8682	0.3027	13.56	0.3027	1.4290	0.3016	1364.2
0.4006	0.8894	0.4006	15.25	0.4006	1.4295	0.4006	1370.8
0.5017	0.9144	0.5017	17.31	0.5017	1.4300	0.5017	1382.2
0.6013	0.9433	0.6013	20.39	0.6013	1.4306	0.6013	1396.3
0.7000	0.9755	0.7000	24.42	0.7000	1.4315	0.7000	1415.5
0.8000	1.0147	0.8000	29.17	0.8000	1.4329	0.8000	1442.2
0.9000	1.0682	0.9000	34.91	0.9000	1.4340	0.9000	1487.5
1.0000	1.1239	1.0000	41.66	1.0000	1.4355	1.0000	1520.5
			(iv) T = 298.15 K, γ-Butyrolactone (1) + 1-Butanol (2)				
0.0000	0.8060	0.0000	17.10	0.0000	1.3973	0.0000	1239.4
0.1000	0.8337	0.1000	18.26	0.1000	1.4014	0.1000	1260.0
0.2004	0.8623	0.2004	19.86	0.1824	1.4048	0.2004	1282.9
0.3002	0.8915	0.3002	21.71	0.2819	1.4082	0.2998	1307.7
0.3979	0.9211	0.3979	23.80	0.3804	1.4117	0.4003	1332.5
0.4999	0.9529	0.5000	26.26	0.5000	1.4159	0.4999	1360.9
0.6000	0.9851	0.6001	29.02	0.6001	1.4195	0.5999	1390.9
0.7001	1.0183	0.7001	32.12	0.7001	1.4233	0.7003	1422.6
0.8001	1.0523	0.7999	35.18	0.7999	1.4273	0.8001	1454.4
0.9000	1.0874	0.9000	38.35	0.9000	1.4312	0.9000	1487.3
1.0000	1.1239	1.0000	41.66	1.0000	1.4355	1.0000	1520.5
			(v) T = 303.15 K, γ-Butyrolactone (1) + 1-Butanol (2)				
0.0000	0.8022	0.0000	16.01	0.0000	1.3965	0.0000	1222.1
0.1000	0.8300	0.1000	17.56	0.1000	1.3997	0.1000	1244.7
0.1824	0.8533	0.2004	19.32	0.1824	1.4027	0.1824	1263.6
0.2819	0.8824	0.3002	21.23	0.2819	1.4062	0.2819	1289.4
0.3804	0.9119	0.3979	23.38	0.3804	1.4097	0.3804	1311.1
0.5000	0.9489	0.4999	25.88	0.5000	1.4140	0.5000	1343.0
0.6001	0.9807	0.6000	28.59	0.6001	1.4182	0.6001	1372.5
0.7001	1.0136	0.7001	31.51	0.7001	1.4227	0.7001	1403.8
0.7999	1.0475	0.8001	34.57	0.7999	1.4258	0.7999	1436.5
0.9000	1.0826	0.9000	37.74	0.9000	1.4296	0.9000	1468.6
1.0000	1.1186	1.0000	41.00	1.0000	1.4331	1.0000	1501.6

$\pm 5 \times 10^{-5}$ g·cm^{-3}. Relative permittivities were measured with a Dipolmeter (model WDW DM-01, Wissenshaftlich Technische Werkstatten GmbH). During the experiments two cells were used, the MFL 3 and MFL 2. The cell MFL 3 is used with a range of permittivity 20 to 50, and the cell MFL 2 is used with a range of permittivity from 7 to 21. A VSI (model 72) thermostat regulated the temperature of the dipolmeter with a temperature stability of ±0.005 K. The reproducibility of the measurements was better than 0.3%. Refractive index measurements were carried out with an Abbe refractometer (Aus JENA, model G), using sodium light (D). The refractometer was thermostated to ±0.005 K with the same thermostat mentioned in relative permittivity. The error in refractive index was better than ±0.0002 units.

The ultrasonic velocity was measured with a DHN model RN-1A sing-around measuring setup, which was described earlier.[11] It consists of a central Polam model CPO 4507-1 controller, a Zopan model kz 2026A-2 universal counter, and a stainless steel cell with two Philips type PXE 4 MHz ultrasound transducers (generator and detector). The cell containing the studied liquid was thermostated by a

Table 3. Coefficients b_j of Eq 1 and the Standard Deviation $\sigma(Y)$

property	b_0	b_1	b_2	b_3	b_4	$\sigma(Y)$
T = 298.15 K, γ-Butyrolactone + Methanol						
ϵ	32.71	27.11	−41.57	37.74	−14.35	0.09
n_D	1.327	0.247	−0.232	0.145	−0.050	0.0012
u (m·s^{-1})	1102.3	770.7	−500.6	148.7		1.20
T = 298.15 K, γ-Butyrolactone + Ethanol						
ϵ	24.55	19.75	−0.21	−2.43		0.026
n_D	1.360	0.115	−0.049	0.010		8×10^{-5}
u (m·s^{-1})	1142.1	537.0	399.5	−220.5		1.60
T = 298.15 K, γ-Butyrolactone + 1-Butanol						
ϵ	17.09	10.69	13.000	6.17	−6.31	0.053
n_D	1.397	0.037	−0.002	0.003		1.8×10^{-4}
u (m·s^{-1})	1239.8	191.7	121.6	−32.3		1.1
T = 303.15 K, γ-Butyrolactone + 1-Butanol						
ϵ	16.01	15.89	8.02	1.17		0.14
n_D	1.397	0.030	0.016	−0.010		2.7×10^{-4}
u (m·s^{-1})	1222.6	206.2	73.9			1.0
T = 298.15 K, γ-Butyrolactone + 1-Octanol						
ϵ	10.17	8.30	2.26	21.17		0.23
n_D	1.428	0.001	0.003	0.003		1.0×10^{-4}
u (m·s^{-1})	1347.0	76.3	−134.4	236.7		0.75

Tamson TMV40 thermostat with temperature stability better than ±0.01 K.

Results and Discussion

The experimental values of densities, relative permittivities, refractive indices, and ultrasound velocities of pure liquids are in very good agreement with the literature values as it is shown in Table 1.

The measured values of density, relative permittivity, refractive index, and ultrasound velocity are reported in Table 2.

The experimental values were fitted to the equation

$$X = \sum_{j=0} b_j x_1^{\,j} \tag{1}$$

where, x_1 is the mole fraction of γ-butyrolactone and b_j are the polynomial coefficients obtained by a least-squares regression. These coefficients are reported in Table 3.

The excess molar volumes, V^E, were calculated using the equation

$$V^E = \sum_i x_i M_i (\rho^{-1} - \rho_i^{-1}) \tag{2}$$

where, ρ and ρ_i are the densities of the mixture and of the pure component i, respectively, and M_i is the molecular mass of component i.

Isentropic compressibilities were calculated from the ultrasound velocities, u, and densities, ρ, using Laplace's relation

$$k_s = u^{-2}\rho^{-1} \tag{3}$$

Another quantity that resulted from the experimental values is the molar polarization, P_m, which has been calculated by the equation

$$P_m = (\epsilon - n_D^{\,2})(2\epsilon - n_D^{\,2})V_m/(9\epsilon) \tag{4}$$

where

$$V_m = (M_1x_1 + M_2x_2)/\rho \tag{5}$$

M_1 and M_2 are the molar masses of the pure components, and ρ is the density of the mixture.

Table 4. Values of Parameters c_j from Eq 9 and the Corresponding Standard Deviations σ from Eq 10

function	c_0	c_1	c_2	c_3	σ
T = 298.15 K, γ-Butyrolactone + Methanol					
V^E (cm^3·mol^{-1})	−2.15	−0.03	−0.68	0.5	0.01
$\Delta\epsilon$	10.1	−3.0	3.7	−5.7	0.05
ΔP_m (cm^3·mol^{-1})	41	−2	19	−52	0.5
Δu (m·s^{-1})	276	−71	18		0.5
Δk_s (10^{-10}m^2·N^{-1})	−4.67	1.19	−0.32		0.003
T = 298.15 K, γ-Butyrolactone + Ethanol					
V^E (cm^3·mol^{-1})	−1.39	−0.14	0.21	0.08	0.004
$\Delta\epsilon$	3.8	1.2			0.02
ΔP_m (cm^3·mol^{-1})	−20.9	22.7			0.3
Δu/(m·s^{-1})	119	0	49	15	0.3
Δk_s (10^{-10}m^2·N^{-1})	−3.95	0.94			0.02
T = 298.15 K, γ-Butyrolactone + 1-Butanol					
V^E (cm^3·mol^{-1})	−0.89	−0.05	0.13	0.23	0.002
$\Delta\epsilon$	12.3	3.2	1.3		0.05
ΔP_m (cm^3·mol^{-1})	−151	78	21		0.9
Δu (m·s^{-1})	−75	18	13		0.3
Δk_s (10^{-10}m^2·N^{-1})	−1.96	0.17	−0.05		0.004
T = 303.15 K, γ-Butyrolactone + 1-Butanol					
V^E (cm^3·mol^{-1})	−1.01	0.19	−0.05	−0.17	0.002
$\Delta\epsilon$	−10.5	1.7	1.7		0.01
ΔP_m (cm^3·mol^{-1})	−115	43	31		0.4
Δu (m·s^{-1})	−75	11	27		0.5
Δk_s (10^{-10}m^2·N^{-1})	−2.07	0.36	−0.36		0.006
T = 298.15 K, γ-Butyrolactone + 1-Octanol					
V^E (cm^3·mol^{-1})	1.73	0.40	0.84	0.71	0.004
$\Delta\epsilon$	34.5	−15	12.7	19	0.34
ΔP_m (cm^3·mol^{-1})	−296	26	253	31	2.1
Δu (m·s^{-1})	−220	−119	−18		0.5
Δk_s (10^{-10}m^2·N^{-1})	−0.05	0.12	−0.03		0.004

The deviations in relative permittivities, ϵ, ultrasound velocities, u, and molar polarization, P_m, are given by the general expression

$$\Delta Y = Y - \sum_i x_i Y_i \tag{6}$$

where Y represents one of the above-mentioned properties for the mixtures and Y_i is the corresponding property of the pure component i.

The deviation in isentropic compressibility, Δk_s, is defined by a similar expression

$$\Delta k_s = k_s - \sum_i \varphi_i k_{s,i} \tag{7}$$

where $k_{s,i}$ is the isentropic compressibility of pure component i and φ_i is its volume fraction

$$\varphi_i = x_iV_i/\sum_i x_iV_i \tag{8}$$

V_i in this equation is the molar volume of pure component i.

The excess functions as well as the property deviations were fitted by a least squares regression to the Redlich and Kister[26] equation

$$V^E \text{ or } \Delta Y = x_1x_2 \sum_{j=0} c_j (x_1 - x_2)^j \tag{9}$$

The standard deviations were calculated with the expression

$$\sigma(V^E) \text{ or } \sigma(\Delta Y) = [\sum_{im} (Y_{exp,i} - Y_{calc,i})^2/(m - n)]^{1/2} \tag{10}$$

where m is the total number of experimental points and n is the number of c_j coefficients.

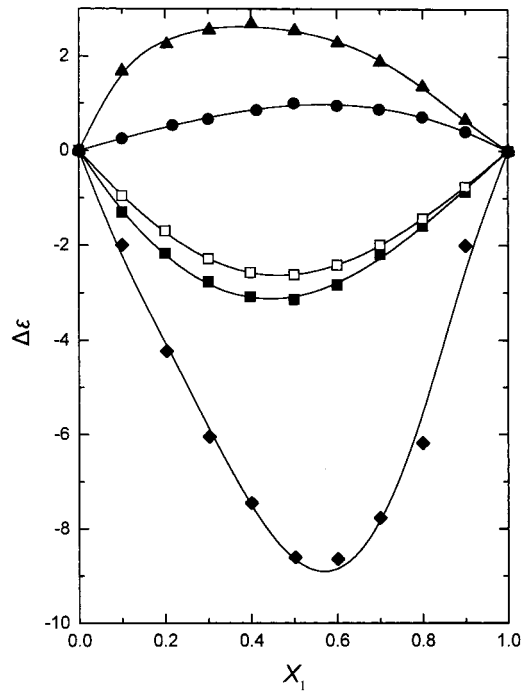

Figure 1. Deviation in relative permittivity, $\Delta\epsilon$, vs mole fraction of γ-butyrolactone, x_1, at 298.15 K: ▲, methanol; ●, ethanol; ■, 1-butanol; □, 1-butanol at 303.15 K; ◆, 1-octanol.

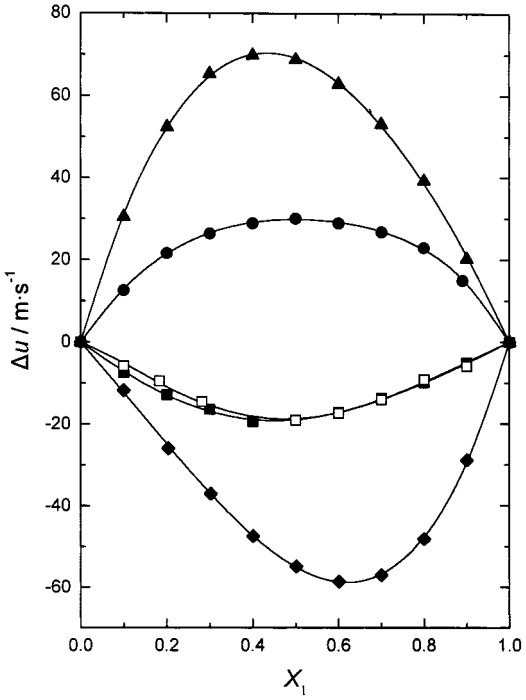

Figure 3. Deviation in ultrasonic velocity, Δu, vs mole fraction of γ-butyrolactone, x_1, at 298.15 K: ▲, methanol; ●, ethanol; ■, 1-butanol; □, 1-butanol at 303.15 K; ◆, 1-octanol.

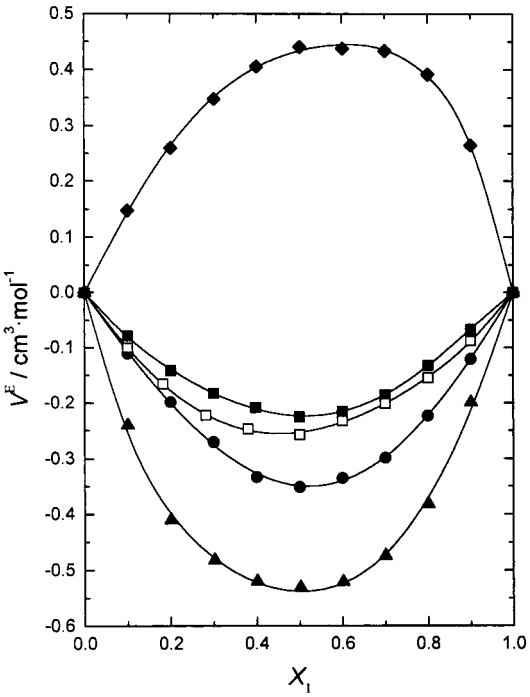

Figure 2. Variation of excess molar volume, V^E, vs mole fraction of γ-butyrolactone, x_1, at 298.15 K: ▲, methanol; ●, ethanol; ■, 1-butanol; □, 1-butanol at 303.15 K; ◆, 1-octanol.

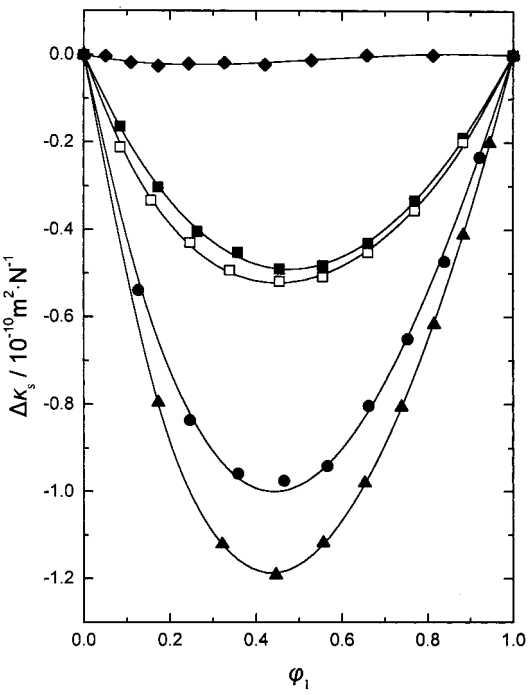

Figure 4. Deviation in isentropic compressibility, Δk_s, vs volume fraction of γ-butyrolactone, φ_1, at 298.15 K: ▲, methanol; ●, ethanol; ■, 1-butanol; □, 1-butanol at 303.15 K; ◆, 1-octanol.

The values of coefficients c_j and the standard deviations σ are reported in Table 4.

The relative permittivity plays an important role in the arrangement of the molecules and in the local structure of the liquid. In addition, the deviation in relative permittivity, $\Delta\epsilon$, reflects the extent of interactions between the dipoles and the hydrogen-bonded complexes. In the case of the binary systems of γ-butyrolactone (γ-BL) with methanol or ethanol, the deviation remains positive through the entire composition range, as shown in Figure 1. The $\Delta\epsilon$ values of ethanol are closer to zero than those of methanol. The $\Delta\epsilon$ values for the remaining systems are always negative through the entire composition range, and the absolute values of $\Delta\epsilon$ extrema increase as the chain length of alcohol increases. The influence of temperature on $\Delta\epsilon$ for the binary systems γ-butyrolactone + 1-butanol is not significant.

The variation of the excess molar volume with mole fraction of γ-butyrolactone is presented in Figure 2. From this figure it is seen that the values of V^E are positive for

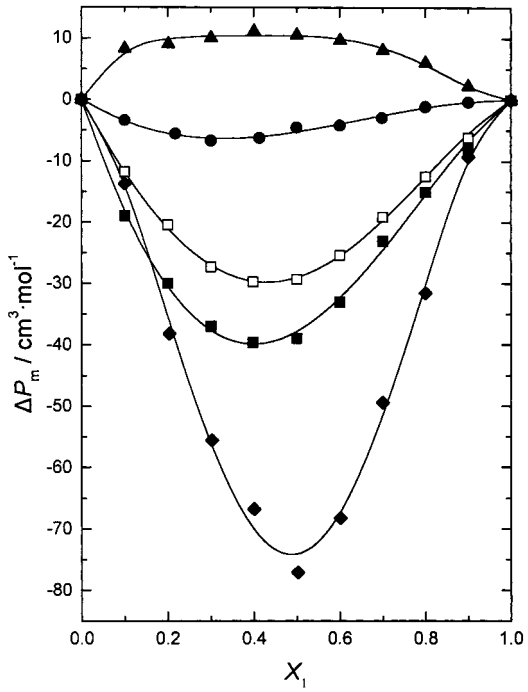

Figure 5. Deviation in molar polarization, P_m, vs mole fraction of γ-butyrolactone, x_1, at 298.15 K: ▲, methanol; ●, ethanol; ■, 1-butanol; □, 1-butanol at 303.15 K; ◆, 1-octanol.

the binary system γ-butyrolactone + 1-octanol but negative for the system γ-butyrolactone with methanol, ethanol, and 1-butanol, while the extrema of the latter curves lie at mole fraction $x = 0.5$. Generally, the values of V^E increase as the carbon chain length of alkanol increases. This is indicative of a more efficient packing as the alkanol chain length decreases, and this is corroborated by the above $\Delta\epsilon$ data. Regarding the influence of temperature on V^E for the binary system γ-butyrolactone + 1-butanol one can detect a slight increase of the minimum as the temperature decreases.

The plots of deviation in ultrasound velocity, Δu, for the binary systems of γ-butyrolactone with methanol, ethanol, 1-butanol (298.5 K), 1-butanol (303.15 K), and 1-octanol are given in Figure 3. The Δu values, as shown in the figure, are negative for the binary systems γ-butyrolactone with 1-octanol and 1-butanol at both temperatures and positive for the system γ-butyrolactone with methanol and ethanol. However, the values of deviation in ultrasound velocity decrease as the carbon chain length of the studied alkanols decreases.

The results of the deviation in isentropic compressibilty, Δk_s, against volume fraction of γ-butyrolactone, φ_1, are represented in Figure 4. The values of Δk_s are negative for all the investigated binary systems through the entire range of volume fraction. The minimum increases according to the following sequence: MeOH < EtOH < BuOH < OctOH. The largest negative value is observed for the system γ-butyrolactone with methanol, while for the system γ-butyrolactone with 1-octanol the Δk_s values are close to zero. The minimum for the studied binary systems remains at $\varphi_1 = 0.45$. The temperature seems to have no significant influence on the deviation in isentropic compressibility for the γ-butyrolactone + 1-butanol system.

The deviation in the molar polarization, ΔP_m, of the studied binary systems vs mole fraction of γ-butyrolactone is plotted in Figure 5. The binary system γ-butyrolactone + methanol gives slightly positive values which are close to zero. The other four systems have negative values through the entire composition range. The curves show a minimum at the $x = 0.5$ mole fraction which increases as the carbon chain length increases.

The above data indicate that the relative strength of specific intermolecular interactions between γ-butyrolactone and 1-alkanols decreases as the alkanol carbon chain length increases. A better assessment of these interactions could be obtained by heat of mixing measurements.

Literature Cited

(1) Pistoia, G., Ed. *Lithium Batteries*; Elsevier: Amsterdam, 1994.
(2) Gabano, J.-P., Ed. *Lithium Batteries;* Academic Press: New York, 1983.
(3) Aurbach, D. The Electrochemical Behavior of Lithium Salt Solutions of γ-butyrolactone with Nobel Metal Electrodes. *J. Electrochem. Soc.* **1989**, *136*, 906−913.
(4) Awwad, A. M.; Salman, M. A.; Hassan, F. A. Liquid−Liquid Equilibria for the Ternary Systems γ-Butyrolactone + *n*-Heptane + Benzene, γ- Butyrolactone + *n*-Heptane + Toluene, and γ-Butyrolactone + *n*-Heptane + *p*-Xylene. *J. Chem. Eng. Data* **1988**, *33*, 263−265.
(5) Ramkumar, D. H. S.; Kudchadker, A. P. Mixture Properties of the Water + γ-Butyrolactone + Tetrahydrofuran System. 1. Densities of γ-Butyrolactone + Water at 303.15−343.15 K and of Tetrahydrofuran + γ-Butyrolactone at 278.15−298.15 K.; Ultrasonic Velocities at 298.15 K for the three Binary Systems Inclusive of Tetrahydrofuran + Water and the Ternary System Tetrahydrofuran + Water + γ-Butyrolactone. *J. Chem. Eng. Data* **1989**, *34*, 459−463.
(6) Côté, J.-F.; Brouillette, D.; Desnoyers, E. J.; Rouleau, J.-F.; St-Arnaud, J.-M.; Perron, G. Dielectric Constants of Acetonitrile, γ-Butyrolactone, Prolylene Carbonate, and 1,2-Dimethoxyethane as a Function of Pressure and Temperature. *J. Solution Chem.* **1996**, *25* (12), 1163−1173.
(7) Avraam, T.; Moumouzias, G.; Ritzoulis, G. A Study on Excess Volumes and Dielectric Properties in the γ-Butyrolactone + *p*-Xylene System at Various Temperatures. *J. Chem. Eng. Data* **1998,** *43*, 51−54.
(8) Fornefeld-Schwarz, M. U.; Svejda, P. Excess Volumes of Liquid Mixtures of Gamma-Butyrolactone (Oxolan-2-one), Gamma-valerolactone (5-Methyloxalan-2-one), Delta-valerolactone (Oxan-2-one), or Epsilon-caprolacton (Oxepan-2-one) + Toluene or + Ethylbenzene at 293.15 and 313.15 K. ELDATA: *Int. Electron. J. Phys.-Chem. Data* **1997**, *3*, 17−24.
(9) Fornefeld-Schwarz, M. U.; Svejda, P. Refractive Indices and Relative Permittivities of Liquid Mixtures of γ-Butyrolactone, γ-Valerolactone, δ-Valerolactone, ϵ-Caprolactone + Benzene, + Toluene, or + Ethylbenzene at 293.15 K and 313.15 K and Atmospheric Pressure. *J. Chem. Eng. Data* **1999**, *44*, 597−604.
(10) Aurbach, D.; Gottlieb, H. The Electrochemical Behavior of Selected Polar Aprotic Systems. *Electrochim. Acta* **1989**, *34*, 141−156.
(11) Papaioannou, D.; Ziakas, D.; Panayiotou, C. Volumetric Properties of Binary Mixtures. 1. 2-Propanone + 2,2,4-Trimethylpentane and *n*-Heptane + Ethanol Mixtures. *J. Chem. Eng. Data* **1991**, *36*, 35−39.
(12) Won, Y. S.; Chang, D. K.; Mills, A. F. Density, Viscosity, Surface Tension, and Carbon Dioxide Solubility and Diffusivity of Methanol, Ethanol, Aqueous Propanol, and Aqueous Ethylene Glycol at 25 °C. *J. Chem. Eng. Data* **1981**, *26*, 140−141.
(13) Aminabhavi, T.; Benerjee, K. Density, Viscosity, Refractive Index, and Speed of Sound in Binary Mixtures of 2-Chloroethanol with Alcanols (C_1−C_6) at 298.15, 303.15, and 308.15. *J. Chem. Eng. Data* **1998**, *43*, 509−513.
(14) Ortega, J.; Matos, J. S. Estimation of Isobaric Expansivities from several Equations of Molar Refraction for Some Pure Organic Compounds. *Mater. Chem. Phys.* **1986**, *15*, 415−425.
(15) Francesconi, R.; Comelli, F. Excess Enthalpies and Excess Volumes of the Liquid Binary Mixtures of Propylene Carbonate + Six Alkanols at 298.15 K. *J. Chem. Eng. Data* **1996**, *41*, 1397−1400.
(16) Riddick, J. A.; Bunger. W. B.; Sakano, T. K. *Organic Solvents*; Wiley: New York, 1986.
(17) Martin, M. C.; Mato, R. B. Isobaric Vapor−Liquid Equilibrium for Methyl Acetate + Methanol + Water at 101.3 kPa. *J. Chem. Eng. Data* **1995**, *40*, 326−327.
(18) El-Banna, M. M.; Radaman, M. S. Density and Viscosity of Octyl, Decyl, and Dodecyl Sodium Sulfates in Ethanol. *J. Chem. Eng. Data* **1995**, *40*, 369−370.
(19) Rauf, M. M.; Stewart, G.; Farhataziz, A. Viscosities and Densities of Binary Mixtures of 1-Alcanols from 15 to 55 °C. *J. Chem. Eng. Data* **1983**, *28*, 324−328.
(20) Artigas, H.; Lafuente, C.; Lopez, M. C.; Royo, F. M.; Urieta, J. S. Isobaric Vapor-Liquid Equilibria for Binary Mixtures of 1-Chlo-

robutane with Isomeric Butanols at 40.0 and 101.3 kPa. *J. Chem. Eng. Data* **1994**, *19*, 729−732.

(21) Rodriguez, A.; Canosa, J.; Orge, B.; Iglesia, M.; Tojo, J. Mixing Properties of the System Methyl Acetate + Methanol + Ethanol at 298.15 K. *J. Chem. Eng. Data* **1996**, *41*, 1446−1449.

(22) Marsh, K. N. *TRC Data Bases for Chemistry and Engineering-TRC Thermodynamic Tables*; Thermodynamic Research Center, Texas A&M University System: College Station, TX, 1994.

(23) Aralaguppi, M. I.; Jadar, C. V.; Aminabhavi, T. M. Density, Viscosity, Refractive Index, and Speed of Sound in Binary Mixtures of Acetonitrile with Methanol, Ethanol, Propan-1-ol, Butan-1-ol, Pentan-1-ol, Hexan-1-ol, Heptan-1-ol, and Butan-2-ol. *J. Chem. Eng. Data* **1999**, *44*, 216−221.

(24) Landolt −Börnstein. *Zahlenwerte und Funktionen. Elektrische Eigenscahften I (Numerical Data and Functional Relationships. Electrical Properties)*; Springer-Verlag: Berlin, 1959.

(25) Arce, A.; Blanco, A.; Soto, A.; Vidal, I. Densities, Refractive Indexes, and Excess Molar Volumes of the Ternary-Systems Water Plus Methanol Plus 1-Octanol and Water Plus Ethanol Plus 1-Octanol and Their Binary Mixtures at 298.15 K. *J. Chem. Eng. Data* **1993**, *38*, 336−340.

(26) Redlich, O.; Kister, A. Algebraic Representation of Thermodynamic Properties and the Classification of Solutions. *J. Phys. Chem*. **1972**, *76*, 2892−2900.

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