Relative Permittivities and Refractive Indices of γ-Butyrolactone with *o*-Xylene and *m*-Xylene

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Relative permittivities and refractive indices of the binary systems γ-butyrolactone + *o*-xylene and γ-butyrolactone + *m*-xylene were measured at (288.15, 293.15, 298.15, 303.15, and 308.15) K. Deviations in relative permittivity, refractive index, molar refraction, and molar polarization were calculated according to several mixing rules. Calculations of apparent dipole moment and dipole moment of the pure liquids were also performed.

Introduction

This work continues our study in mixtures of γ-butyrolactone with xylenes. The densities and viscosities of γ-butyrolactone + *o*-xylene and γ-butyrolactone + *m*-xylene mixtures are reported elsewhere (Moumouzias et al., in press), whereas the γ-butyrolactone + *p*-xylene system was studied previously (Avraam et al., 1998). Here, the relative permittivities and the refractive indices of γ-butyrolactone + *o*-xylene and γ-butyrolactone + *m*-xylene mixtures were measured.

A number of mixing rules have been proposed in the literature for the relative permittivity, the refractive index, and the molar refraction of a mixture. Some of them are supported by a theory, and some are simple empirical rules. One purpose of this paper is to examine the applicability of the more common mixing rules to the studied systems.

Experimental Section

Relative permittivity measurements were made by means of a dipolmeter of Wissenschaftlich Technische Werkstätten GmbH, model DM-01. The uncertainty in the values was estimated to be no more than 0.3%. Refractive indices were measured with an Abbe refractometer aus JENA, model G, and the values were obtained for Na D light with an error lower than ±0.0002 units. The temperature in the dipolmeter and the refractometer was regulated with a YSI (model 72) thermostat with a ±0.005 K stability. Details about calibration of the apparatuses are given elsewhere (Moumouzias and Ritzoulis, 1997).

o-Xylene, *m*-xylene (Fluka, puriss p.a. >99%), and γ-butyrolactone (Aldrich, >99%) were purified by distillation. All solutions were prepared by mass using a Shimadzu AEG 220 analytical balance accurate to four decimal places, whereas the atomic masses were taken from the atomic mass table of IUPAC (IUPAC, 1996). The uncertainty in mole fraction was <±0.00003.

Results and Discussion

The experimental data for the two systems are presented in Tables 1 and 2. Very few literature values were found for the pure components and are presented with the experimental ones in Table 3, for 298.15 K. For the refractive indexes of *o*-xylene and *m*-xylene we obtained the experimental values 1.5028 and 1.4945, which are in

Table 1. Experimental Values of Relative Permittivity ϵ, Refractive Index n_D, and Density ρ for γ-Butyrolactone (1) + *o*-Xylene Mixtures[a]

x_1	ϵ				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
0.0000	2.6	2.58	2.56	2.55	2.53
0.1199	4.86	4.80	4.71	4.60	4.53
0.1801	6.12	6.00	5.88	5.78	5.67
0.2998	9.17	8.94	8.77	8.67	8.49
0.4005	12.34	12.05	11.80	11.61	11.42
0.5044	16.11	15.75	15.46	15.17	14.97
0.5994	20.02	19.59	19.28	18.95	18.66
0.7011	24.73	24.28	23.96	23.58	23.15
0.7885	29.33	28.91	28.56	28.07	27.55
0.9087	36.66	36.27	35.77	35.07	34.52
1.0000	43.07	42.35	41.68	41.06	40.48

x_1	n_D				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
0.0000	1.5079	1.5053	1.5028	1.5003	1.4978
0.1199	1.5033	1.5010	1.4983	1.4960	1.4934
0.1801	1.5007	1.4984	1.4957	1.4935	1.4909
0.2998	1.4949	1.4925	1.4901	1.4879	1.4853
0.4005	1.4894	1.4869	1.4846	1.4825	1.4800
0.5044	1.4830	1.4805	1.4784	1.4763	1.4739
0.5994	1.4766	1.4741	1.4720	1.4700	1.4677
0.7011	1.4689	1.4665	1.4646	1.4624	1.4602
0.7885	1.4614	1.4591	1.4571	1.4551	1.4530
0.9087	1.4495	1.4476	1.4457	1.4436	1.4416
1.0000	1.4392	1.4374	1.4358	1.4336	1.4316

x_1	ρ/g cm^{-3}				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
0.0000	0.8853	0.8808	0.8762	0.8715	0.8671
0.1199	0.9055	0.9010	0.8967	0.8920	0.8877
0.1801	0.9167	0.9122	0.9078	0.9032	0.8989
0.2998	0.9406	0.9363	0.9318	0.9270	0.9226
0.4005	0.9625	0.9582	0.9536	0.9488	0.9443
0.5044	0.9868	0.9824	0.9778	0.9729	0.9683
0.5994	1.0105	1.0062	1.0015	0.9967	0.9919
0.7011	1.0379	1.0334	1.0288	1.0240	1.0191
0.7885	1.0632	1.0587	1.0540	1.0493	1.0443
0.9087	1.1013	1.0968	1.0921	1.0873	1.0821
1.0000	1.1333	1.1288	1.1239	1.1188	1.1135

[a] Density values were taken from Moumouzias et al., in press, whereas the values of pure γ-butyrolactone were from Avraam et al., 1998.

good agreement with the values reported by Serrano et al. (1990), by Tanaka et al. (1975), by Tojo and Diaz (1995),

Table 2. Experimental Values of Relative Permittivity ϵ, Refractive Index n_D, and Density ρ, for γ-Butyrolactone (1) + m-Xylene Mixtures[a]

x_1	ϵ				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
0.0000	2.43	2.41	2.39	2.37	2.35
0.1012	4.13	4.08	4.04	4.00	3.97
0.2140	6.54	6.45	6.36	6.28	6.18
0.3091	9.04	8.89	8.74	8.61	8.50
0.4022	11.93	11.72	11.50	11.31	11.17
0.5343	16.85	16.54	16.25	15.96	15.71
0.5955	19.48	19.11	18.80	18.46	18.15
0.7101	24.99	24.56	24.21	23.78	23.41
0.8371	32.20	31.62	31.19	30.68	30.16
0.9027	36.35	35.71	35.20	34.66	34.07
1.0000	43.07	42.35	41.68	41.06	40.48

x_1	n_D				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
0.0000	1.4997	1.4973	1.4945	1.4920	1.4896
0.1012	1.4970	1.4946	1.4920	1.4896	1.4871
0.2140	1.4930	1.4907	1.4882	1.4858	1.4831
0.3091	1.4889	1.4867	1.4843	1.4820	1.4792
0.4022	1.4845	1.4823	1.4800	1.4776	1.4750
0.5343	1.4771	1.4750	1.4727	1.4704	1.4682
0.5955	1.4733	1.4712	1.4690	1.4666	1.4646
0.7101	1.4654	1.4634	1.4613	1.4588	1.4569
0.8371	1.4552	1.4533	1.4513	1.4488	1.4468
0.9027	1.4493	1.4474	1.4454	1.4430	1.4410
1.0000	1.4392	1.4374	1.4358	1.4336	1.4316

x_1	ρ/g cm^{-3}				
	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
0.0000	0.8684	0.8642	0.8602	0.8553	0.8521
0.1012	0.8872	0.8831	0.8790	0.8742	0.8709
0.2140	0.9097	0.9056	0.9014	0.8965	0.8931
0.3091	0.9299	0.9258	0.9216	0.9166	0.9131
0.4022	0.9511	0.9469	0.9427	0.9377	0.9340
0.5343	0.9839	0.9796	0.9753	0.9702	0.9663
0.5955	1.0003	0.9960	0.9917	0.9865	0.9825
0.7101	1.0334	1.0290	1.0246	1.0194	1.0152
0.8371	1.0741	1.0696	1.0651	1.0599	1.0552
0.9027	1.0969	1.0924	1.0877	1.0826	1.0776
1.0000	1.1333	1.1288	1.1239	1.1188	1.1135

[a] Density values were taken from Moumouzias et al., in press, whereas the values of pure γ-butyrolactone were from Avraam et al., 1998.

**Table 3. Physical Properties of the Pure Components at 298.15 K**

	γ-butyrolactone	o-xylene	m-xylene
refractive index	1.4358[a]	1.5028[b]	1.4945[b]
	1.43477[c]	1.50268[d]	1.49466[d]
		1.50252[e]	1.49416[e]
		1.50177[f]	1.49443[f]
			1.4945[g]
relative permittivity	41.68[a]	2.56[b]	2.39[b]
	41.7[h]		
	39.1[h]		
	41.95[i]		

[a] Avraam et al., 1998. [b] This work. [c] Awwad et al. [d] Serrano et al., 1990. [e] Tanaka et al., 1975. [f] Tojo and Diaz, 1995. [g] Aralaguppi et al., 1992. [h] Pistoia, 1994. [i] D'Aprano et al., 1983.

and by Aralaguppi et al. (1992). The difference between the value given by Awwad et al. (1988) for the refractive index of γ-butyrolactone and our value is $\sim$0.07%. Agreement is observed in relative permittivity values of γ-butyrolactone, excepting the value 39.1 reported by Pistoia (1994).

Deviations in relative permittivity $\Delta\epsilon$ were calculated from the most usual expressions

$$\Delta\epsilon_x = \epsilon - (x_1\epsilon_1 + x_2\epsilon_2) \tag{1}$$
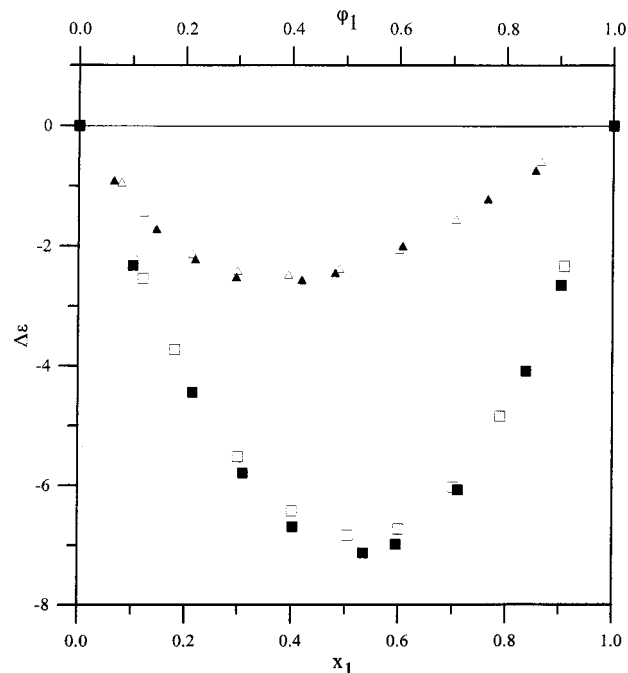
Figure 1. Variation of $\Delta\epsilon$ with the mole fraction x_1 (squares, eq 1) or the volume fraction φ_1 (triangles, eq 2) of γ-butyrolactone, at 298.15 K: open symbols, γ-butyrolactone + o-xylene system; full symbols, γ-butyrolactone + m-xylene system.

(Payne and Theodorou, 1972; Nath and Chaudhary, 1992; Seshadri and Subrahmanyan, 1990; Ruostesuo and Liias-Lepisto, 1991; Corradini et al., 1992) and

$$\Delta\epsilon_\varphi = \epsilon - (\varphi_1\epsilon_1 + \varphi_2\epsilon_2) \tag{2}$$

(Böttcher, 1973; Aralaguppi et al., 1991; Buep and Barün, 1988; Nath and Narain, 1982; Jannelli et al., 1983). In the above equations, ϵ is the relative permittivity of the mixture, ϵ_i is the relative permittivity of the pure components, x_i is the mole fraction, and φ_i the volume fraction.

The dependence of $\Delta\epsilon_x$ and $\Delta\epsilon_\varphi$ on mole and volume fraction is shown at 298.15 K, in Figure 1. The temperature effect on $\Delta\epsilon_x$ and $\Delta\epsilon_\varphi$ is insignificant. Both deviations were negative, but the $\Delta\epsilon_x$ values are about three times higher than those of $\Delta\epsilon_\varphi$. This could be attributed to the fact that eq 2 was proposed for mixtures involving one or both highly polar components (Jannelli et al., 1983).

Several mixing rules are available for the refractive index of a mixture (Heller, 1965). Some of them are not suitable when there is a large change of volume on mixing, whereas the most common are (Tasic et al., 1992; Aminabhavi, 1984; Tojo and Diaz, 1995) the Lorentz–Lorentz relation

$$\frac{n_D^2 - 1}{n_D^2 + 2} = \varphi_1\left(\frac{n_{D,1}^2 - 1}{n_{D,1}^2 + 2}\right) + \varphi_2\left(\frac{n_{D,2}^2 - 1}{n_{D,2}^2 + 2}\right) \tag{3}$$
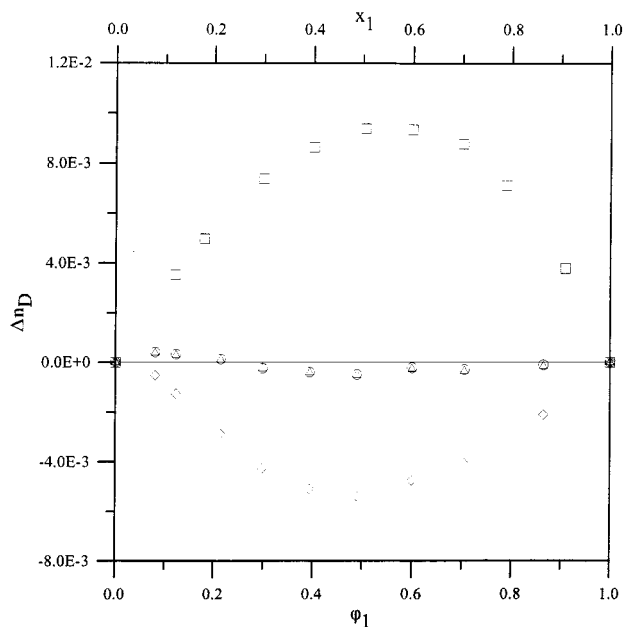
the Gladstone–Dale relation

$$n_D - 1 = \varphi_1(n_{D,1} - 1) + \varphi_2(n_{D,2} - 1) \tag{4}$$

and the Arago–Biot relation

$$n_D = \varphi_1 n_{D,1} + \varphi_2 n_{D,2} \tag{5}$$

Another equation which has been used (Nakata and

Figure 2. Variation of Δn_D, with the mole fraction x_1 ($\square$, eq 6) or the volume fraction φ_1 ($\bigcirc$, eq 3; $\triangle$, eq 4; $\diamond$, eq 5) of γ-butyrolactone, for the γ-butyrolactone + o-xylene system, at 298.15 K.

Figure 3. Variation of Δn_D, with the mole fraction x_1 ($\square$, eq 6) or the volume fraction φ_1 ($\bigcirc$, eq 3; $\triangle$, eq 4; $\diamond$, eq 5) of γ-butyrolactone, for the γ-butyrolactone + m-xylene system, at 298.15 K.

Sakurai, 1987; Iglesias et al., 1996) is the simple additive relation

$$n_D = x_1 n_{D,1} + x_2 n_{D,2} \tag{6}$$

In the above relations, n_D is the refractive index of the mixture and $n_{D,i}$ is the refractive index of the pure components.

To illustrate the predictive abilities of the previous mixing rules, the deviations between the experimental and the calculated values of the refractive indices Δn_D were calculated according to

$$\Delta n_D = n_{D,exp} - n_{D,cal} \tag{7}$$

where $n_{D,cal}$ represents the refractive index calculated each time from eqs 3–6. The deviations Δn_D are almost independent of temperature. Therefore, only the dependence of Δn_D on mole or volume fraction of γ-butyrolactone, at 298.15 K, is presented graphically in Figure 2 for the γ-butyrolactone + o-xylene system and in Figure 3 for the γ-butyrolactone + m-xylene system. Both systems show about the same behavior; Δn_D according to eq 6 was positive with a maximum at $x_1 \sim 0.5$, whereas Δn_D (eq 5) was negative with a minimum at $\varphi_1 \sim 0.5$. Δn_D (eqs 3 and 4) values were very close to zero, indicating that Lorentz–Lorentz and Gladstone–Dale relations predict very well the refractive indices of the studied systems.

The molar refraction can be calculated from refractive index data, from a large number of relations, which have been proposed. The relations which are used most often in modern literature are the Lorentz–Lorentz relation (L–L)

$$R_m = \frac{n_D^2 - 1}{n_D^2 + 2} V \tag{8}$$

the Gladstone–Dale (G–D) relation

$$R_m = (n_D - 1) V \tag{9}$$
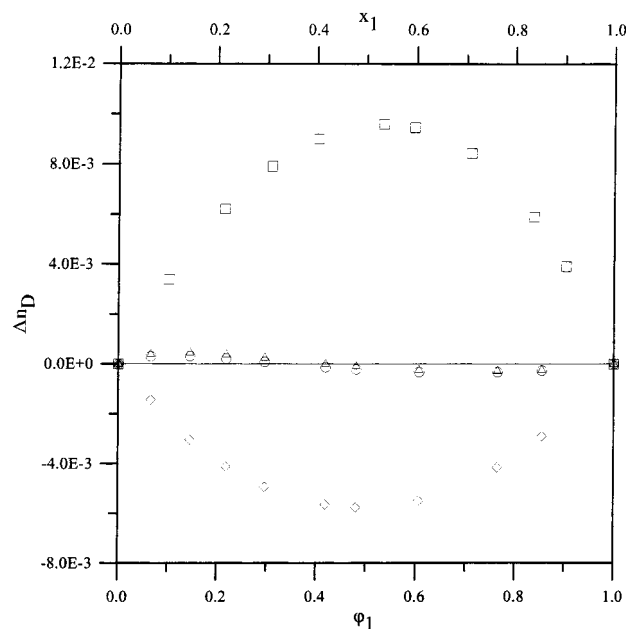
the Eykman (Eyk) relation

$$R_m = \frac{n_D^2 - 1}{n_D + 0.4} V \tag{10}$$

the Newton (N) relation

$$R_m = (n_D^2 - 1) V \tag{11}$$

and the Oster (Ost) relation

$$R_m = \frac{(n_D^2 - 1)(2n_D^2 + 1)}{n_D^2} V \tag{12}$$

(Abdel-Azim and Munk, 1987), where V is the molar volume. The Lorentz–Lorentz rule is usually preferred because it is supported by a plausible theory. However, the remaining rules seem to be as good as the Lorentz–Lorentz one (Abdel-Azim and Munk, 1987; Aralaguppi et al., 1991). The deviation in molar refraction ΔR_m was calculated from the common expressions

$$\Delta R_{m,x} = R_m - (x_1 R_{m,1} + x_2 R_{m,2}) \tag{13}$$

(Tojo and Diaz, 1995) and

$$\Delta R_{m,\varphi} = R_m - (\varphi_1 R_{m,1} + \varphi_2 R_{m,2}) \tag{14}$$

(Aralaguppi et al., 1991; Aminabhavi, 1993). In the above equations R_m is the molar refraction of the mixture and $R_{m,i}$ is the molar refraction of the pure components.

The dependence of ΔR_m on temperature was found to be neglible and $\Delta R_{m,\varphi}-\varphi_1$ plots for 298.15 K are given in Figures 4 and 5 together with $\Delta R_{m,x} - x_1$ plots for Lorentz–Lorentz and Oster relations. For both systems $\Delta R_{m,\varphi}$ values were negative, following a smoothing trend with minimums at $\varphi_1 \sim 0.45$–0.5. $\Delta R_{m,\varphi}$ absolute values were also in the order $\Delta R_{m,\varphi}$ (L–L) $< \Delta R_{m,\varphi}$(G–D) $< \Delta R_{m,\varphi}$(Eyk) $< \Delta R_{m,\varphi}$(N) $< \Delta R_{m,\varphi}$(Ost). Analogous behavior was observed in
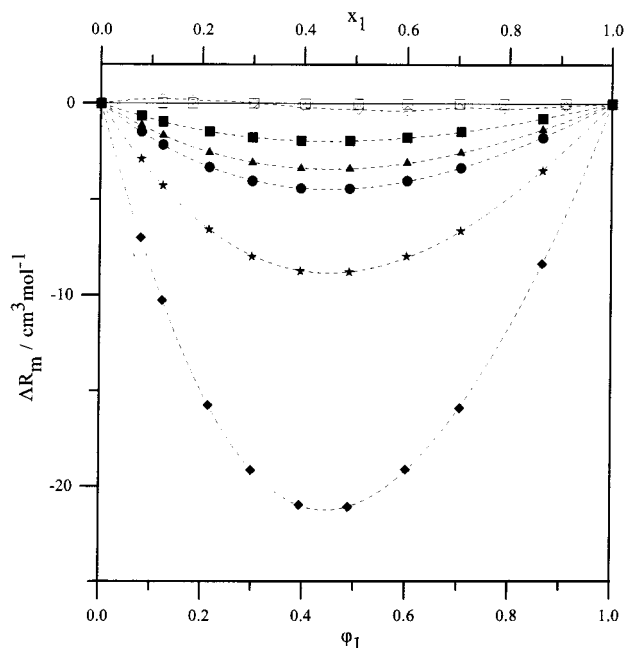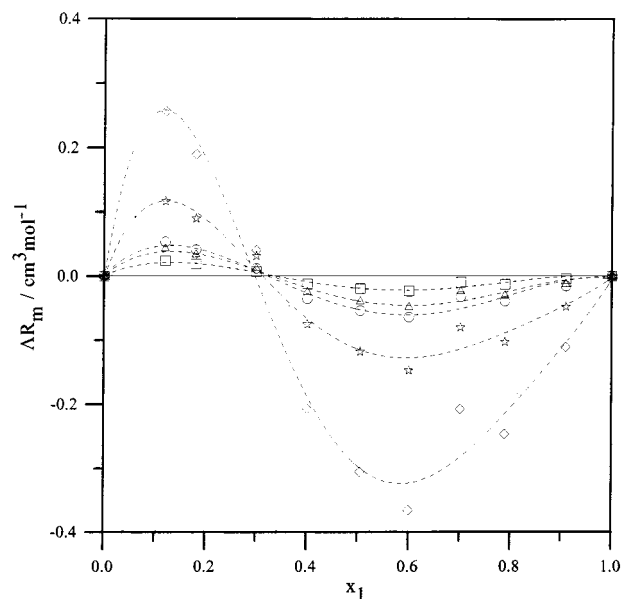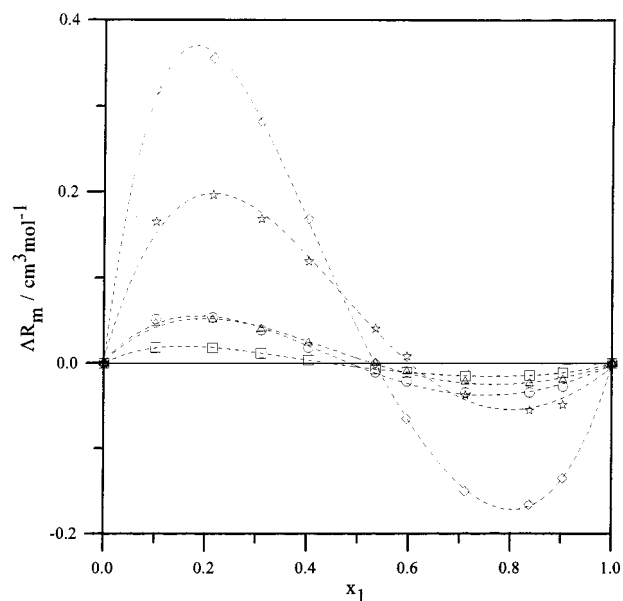
Figure 4. Variation of ΔR_m with the mole fraction x_1 or the volume fraction φ_1 of γ-butyrolactone, for the γ-butyrolactone + o-xylene system, at 298.15 K. Relations based on x_1: □, Lorentz−Lorentz; ◇, Oster. Relations based on φ_1: ■, Lorentz−Lorentz; ▲, Gladstone−Dale; ●, Eykman; ★, Newton; ◆, Oster. Curves show the general trend.
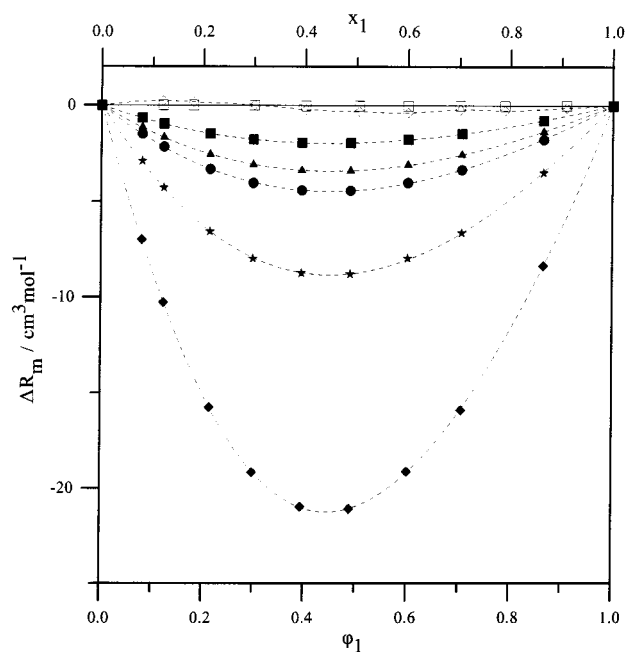
Figure 5. Variation of ΔR_m with the mole fraction x_1 or the volume fraction φ_1 of γ-butyrolactone, for the γ-butyrolactone + m-xylene system, at 298.15 K. Relations based on x_1: □, Lorentz−Lorentz; ◇, Oster. Relations based on φ_1: ■, Lorentz−Lorentz; ▲, Gladstone−Dale; ●, Eykman; ★, Newton; ◆, Oster. Curves show the general trend.

methyl acetoacetate + mesitylene (Aralaguppi et al., 1992) as well as in bromoform + n-hexadecane (Aralaguppi et al., 1991) mixtures. The $\Delta R_{m,x}$ values were very close to zero, and in order to have a better estimation, $\Delta R_{m,x}-x_1$ plots are presented in Figures 6 and 7. As one can see, for both systems, $\Delta R_{m,x}-x_1$ plots follow a S-shape behavior in the order $\Delta R_{m,x}(L-L) < \Delta R_{m,x}(G-D) < \Delta R_{m,x}(Eyk) < \Delta R_{m,x}(N) < \Delta R_{m,x}(Ost)$, indicating that the Lorentz−Lorentz relation gives the better results.

Figure 6. Variation of ΔR_m with the mole fraction x_1 of γ-butyrolactone, for the γ-butyrolactone + o-xylene system, at 298.15 K. Relations: □, Lorentz−Lorentz; △, Gladstone−Dale; ○, Eykman; ☆, Newton; ◇, Oster. Curves show the general trend.

Figure 7. Variation of ΔR_m with the mole fraction x_1 of γ-butyrolactone, for the γ-butyrolactone + m-xylene system, at 298.15 K. Relations: □, Lorentz−Lorentz; △, Gladstone−Dale; ○, Eykman; ☆, Newton; ◇, Oster. Curves show the general trend.

The molar polarization P_m was calculated from the Kirkwood−Frohlich equation

$$P_m = \frac{(\epsilon - n_D{}^2)(2\epsilon + n_D{}^2)}{9\epsilon}V \qquad (15)$$

(Nath and Tripathi, 1984; Ruostesuo and Liias-Lepisto, 1991), and deviations of the molar polarization ΔP_m were calculated using

$$\Delta P_m = P_m - (x_1 P_{m,1} + x_2 P_{m,2}) \qquad (16)$$

(Ruostesuo and Liias-Lepisto, 1991). In eq 16 $P_{m,i}$ are the molar polarizations of the pure components. The dependence of ΔP_m on temperature was also found to be insig-
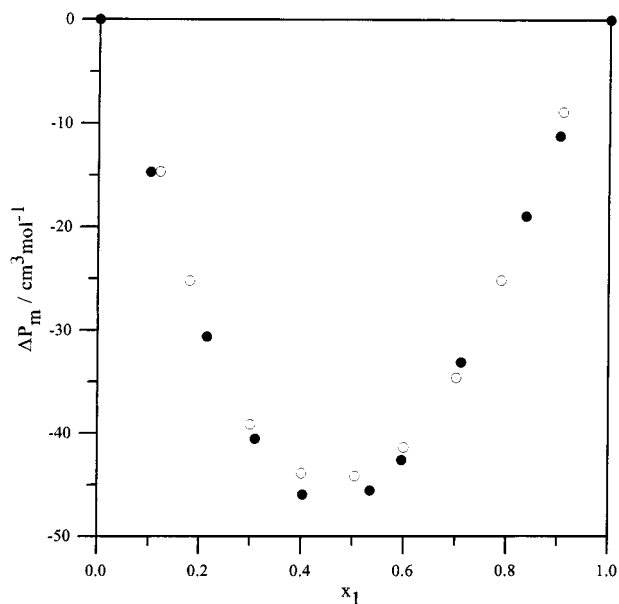
Figure 8. Variation of ΔP_m with the mole fraction of γ-butyrolactone x_1 at 298.15 K: $\bigcirc$, γ-butyrolactone + o-xylene system; $\bullet$, γ-butyrolactone + m-xylene system.

Figure 9. Variation of $\langle u \rangle$ with the mole fraction of γ-butyrolactone x_1 at 298.15 K: $\bigcirc$, γ-butyrolactone + o-xylene system; $\bullet$, γ-butyrolactone + m-xylene system.

nificant and deviations of the molar polarization against mole fraction of γ-butyrolactone at 298.15 are plotted in Figure 8. As can be seen, the two studying systems have the same behavior, showing negative ΔP_m values with a minimum at $x_1 \sim 0.5$. The same behavior has also been observed in the γ-butyrolactone + p-xylene system (Avraam et al., 1998).

The dipole moments of the pure liquids μ_1 were calculated from the expression

$$\mu_1{}^2 = \frac{9kTV(2\epsilon + \epsilon_\infty)(\epsilon - \epsilon_\infty)}{4\pi N_A \epsilon (\epsilon_\infty + 2)^2} \tag{17}$$

(Böttcher, 1973; Minkin et al., 1970) and found at 298.15 K to be 4.3 D, 0.53 D, and 0.39 D, for γ-butyrolactone, o-xylene, and m-xylene, correspondingly. These values are in agreement with the literature ones: 4.19 D for γ-butyrolactone, 0.54 D for o-xylene, and 0.35 D for m-xylene (McClelan, 1989). In the above equation k is the Boltzman's constant, T is the absolute temperature, N_A is Avogadro's number, and ϵ_∞ is the high-frequency relative permittivity, taken equal to the square of the refractive index.

The dipole moment values of o-xylene and m-xylene are not zero but are much lower than the dipole moment value of γ-butyrolactone. In this case the apparent dipole moment of γ-butyrolactone, $\langle u \rangle$ could be calculated from the Kirkwood–Fröhlich equation

$$\langle u \rangle^2 = \frac{9kT(2\epsilon + \epsilon_{\infty,1})^2}{4\pi N_A x_1(\epsilon_{\infty,1} + 2)^2(2\epsilon + 1)} \left[\frac{V(\epsilon - 1)}{\epsilon} - \frac{3(1 - x_1)V_2(\epsilon_2 - 1)}{(2\epsilon + \epsilon_2)} - \frac{3x_1 V_1(\epsilon_{\infty,1} - 1)}{(2\epsilon + \epsilon_{\infty,1})} \right] \tag{18}$$

which has been proposed for polar solutes in nonpolar solvents (Böttcher, 1973). The indices 1 and 2 in eq 18 correspond to polar solute (γ-butyrolactone) and to nonpolar solvent (o-xylene or m-xylene). Variation of $\langle u \rangle$ with the mole fraction of γ-butyrolactone x_1 at 298.15 K is presented for the two systems in Figure 9. The $\langle u \rangle$ value for the γ-butyrolactone + o-xylene system is reduced from $x_1 \approx 0.1$
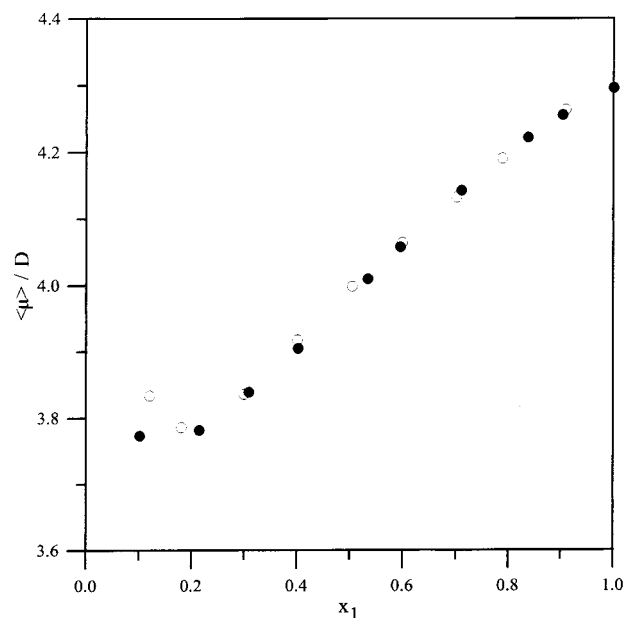
to 0.2 and is increased gently after $x_1 \approx 0.2$. Similar behavior has been observed in other studied systems (Stokes and Marsh, 1976; French et al., 1988). The dipole moments of the complex species that are formed in this region seem to be orientated in an antiparallel fashion. A change of the slope of the $\langle u \rangle$–x_1 curve of the γ-butyrolactone + m-xylene system is observed at $x_1 \approx 0.2$, showing an increment of $\langle u \rangle$ values after this concentration, as for the γ-butyrolactone + o-xylene system. As there are no measurements for $x_1 < 0.1$, for both systems, we are not able to draw a conclusion for this region.

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