

A Study in Mixtures of γ -Butyrolactone with *o*-Xylene and *m*-Xylene: Densities and Viscosities

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The densities in the temperature range 288.15 K to 308.15 K and the viscosities in the temperature range 293.15 K to 308.15 K of the binary systems γ -butyrolactone + *o*-xylene and *m*-xylene were measured. Viscosity data were correlated by the Heric, McAllister, and Hind–McLaughlin–Ubbelohde equations. Viscosity deviations from mole fraction linearity and excess volumes were calculated and found to be negative. Excess volumes were also fitted to a Redlich–Kister type equation.

Introduction

The physical properties and the electrochemical behavior of γ -butyrolactone are of great interest because this solvent is associated with lithium batteries (Gabano, 1983; Pistoia, 1994; Aurbach, 1989). γ -Butyrolactone has high relative permittivity (41.7 at 298.15 K; Pistoia, 1994) and viscosity (1.738 mPa s at 298.15 K; Ramkumar and Kudchadker, 1989b), as well. Since the conductivity of an electrolytic system can be improved by mixing substances of high relative permittivity with substances of low viscosity, mixtures of γ -butyrolactone with low viscosity substances attract a lot of interest (Tobishima and Okada, 1985).

This paper is a part of a general study of the physico-chemical properties of γ -butyrolactone binary mixtures with xylenes. In a previous paper (Avraam et al., 1998) the densities, the excess volumes, and some dielectric properties of mixtures of γ -butyrolactone with *p*-xylene were examined. Here, we continue this study by measuring the densities and the viscosities of the mixtures of γ -butyrolactone with *o*-xylene and *m*-xylene.

Experimental Section

o-Xylene and *m*-xylene (Fluka, puriss p.a. >99%) were distilled, and the middle fractions were collected. γ -Butyrolactone (Aldrich, >99%) was also distilled under reduced pressure. The solutions were prepared by mass with a Shimadzu, AEG 220 analytical balance of four decimal places. Special precautions to avoid contact of the solutions with the atmosphere before the measurements were not taken. The atomic masses were taken from the atomic mass Table of IUPAC (IUPAC, 1996), and the error in the mole fraction was estimated to be lower than ± 0.00003 .

An Anton-Paar DMA (measuring cell 602 and processing unit 60) vibrating tube densimeter was used for density measurements. The densimeter was calibrated with air and double-distilled water, and thermostated by a Haake F3-K digital thermostat. The accuracy in density measurements was better than $5 \times 10^{-5} \text{ g cm}^{-3}$.

Kinematic viscosities were determined using Ubbelohde suspended-level viscometers. Flow times were measured to 0.01 s with an electronic stopwatch. Kinematic viscosity, ν , was evaluated from flow time, t , by

$$\nu = K(t - E/t^2)$$

Table 1. Experimental Values of Density, ρ , and Viscosity, η , for γ -Butyrolactone (1) + *o*-Xylene Mixtures

x_1	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
	$\rho/(\text{g cm}^{-3})$				
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
	$\eta/(\text{mPa s})$				
0.0000		0.812	0.758	0.711	0.665
0.1199		0.907	0.854	0.795	0.747
0.1801		0.955	0.901	0.839	0.787
0.2998		1.054	0.996	0.927	0.867
0.4005		1.145	1.081	1.006	0.937
0.5044		1.248	1.175	1.093	1.014
0.5994		1.351	1.269	1.178	1.090
0.7011		1.472	1.379	1.276	1.179
0.7885		1.584	1.480	1.367	1.263
0.9087		1.750	1.629	1.502	1.390
1.0000		1.883	1.745	1.612	1.498

where K and E are constants and the term E/t^2 is the kinetic energy factor (JENA^{ER} Glaswerk Schott & Gen. Mainz). The calibration of the viscometers is reported elsewhere (Moumouzias and Ritzoulis, 1992). The temperature was regulated by a Yellow Springs Instrument Co. (model 72) thermostat with a stability of ± 0.005 K. At least four measurements were taken for each composition and temperature, and the results were averaged. Viscosity, η , was obtained from kinematic viscosity, ν , and density, ρ , by

$$\eta = \nu\rho$$

The reproducibility in viscosity was better than 0.2%.

Results and Discussion

The experimental values of density and viscosity for γ -butyrolactone + *o*-xylene and γ -butyrolactone + *m*-xylene system are given in Tables 1 and 2, respectively. The

Table 2. Experimental Values of Density, ρ , and Viscosity, η , for γ -butyrolactone (1) + *m*-Xylene Mixtures

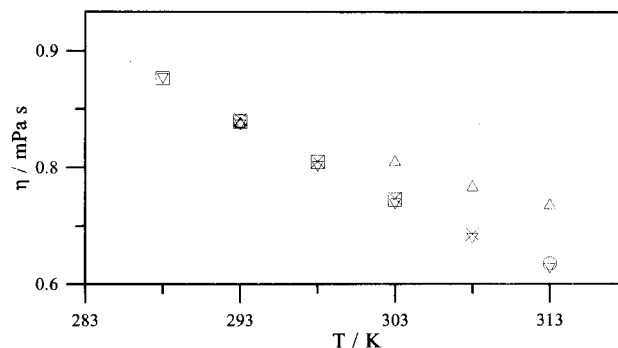
x_1	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
	$\rho/(\text{g cm}^{-3})$				
0.0000	0.8684	0.8642	0.8602	0.8553	0.8521
0.1012	0.8872	0.8831	0.8790	0.8742	0.8709
0.214	0.9097	0.9056	0.9014	0.8965	0.8931
0.3091	0.9299	0.9258	0.9216	0.9166	0.9131
0.4022	0.9511	0.9469	0.9427	0.9377	0.9340
0.5343	0.9839	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
	$\eta/(\text{mPa s})$				
0.0000		0.618	0.588	0.555	0.527
0.1012		0.690	0.648	0.610	0.573
0.214		0.777	0.725	0.679	0.636
0.3091		0.859	0.798	0.752	0.692
0.4022		0.948	0.880	0.829	0.761
0.5343		1.093	1.016	0.951	0.875
0.5955		1.169	1.088	1.013	0.936
0.7101		1.332	1.235	1.147	1.060
0.8371		1.544	1.437	1.325	1.226
0.9027		1.671	1.552	1.431	1.323
1.0000		1.883	1.745	1.612	1.498

Table 3. Physical Properties of the Pure Components

	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
	$\rho/(\text{g cm}^{-3})$				
γ -butyrolactone	1.1333 ^a 1.1336 ^b	1.1288 ^a 1.1280 ^b 1.12917 ^c	1.1239 ^a 1.1237 ^b 1.12421 ^d	1.1188 ^a 1.1189 ^b	1.1135 ^a
<i>o</i> -xylene	0.8853 ^a 0.88288 ^e	0.8808 ^a 0.87946 ^e 0.877 ^f	0.8762 ^a 0.87558 ^e 0.87563 ^g	0.8715 ^a 0.87136 ^e 0.856 ^f	0.8671 ^a 0.855 ^f
<i>m</i> -xylene	0.8684 ^a 0.86809 ^e	0.8642 ^a 0.86378 ^e 0.8641 ^g	0.8602 ^a 0.86006 ^e 0.85986 ^g 0.8596 ^h	0.8553 ^a 0.85587 ^e 0.85561 ^g 0.8554 ⁱ	0.8521 ^a
	$\eta/(\text{mPa s})$				
γ -butyrolactone		1.883 ^a	1.745 ^a 1.738 ^j	1.612 ^a 1.6097 ^j	1.498 ^a
<i>o</i> -xylene		0.812 ^a 0.809 ^e 0.806 ^f 0.807 ^g 0.8102 ^k	0.758 ^a 0.757 ^e 0.754 ^g	0.711 ^a 0.709 ^e 0.757 ^f 0.706 ^g	0.665 ^a 0.725 ^f 0.662 ^g
<i>m</i> -xylene		0.618 ^a 0.621 ^e 0.615 ^g 0.620 ^k	0.588 ^a 0.590 ^e 0.579 ^g	0.555 ^a 0.556 ^e 0.547 ^g	0.527 ^a 0.517 ^g

^a Experimental values. ^b Ramkumar and Kudchadker, 1989a. ^c Klein and Svejda, 1995. ^d Awwad et al., 1988. ^e Serrano et al., 1990. ^f Swain et al., 1997. ^g TRC, 1998. ^h Chang and Lee, 1995. ⁱ Ruiz et al., 1989. ^j Ramkumar and Kudchadker, 1989b. ^k Lange, 1956.

experimental and the literature values of the pure components are presented in Table 3. The experimental values of γ -butyrolactone and *m*-xylene are in very good agreement with the literature ones. The agreement is better than 0.1% for density and 0.5% for viscosity data, for both substances. Agreement is also observed for *o*-xylene, but the experimental data at 303.15 and 308.15 K do not agree very well with the data reported by Swain et al. (1997). So, the viscosity data of pure *o*-xylene were plotted against temperature in Figure 1. As can be seen, our values follow the same linear trend with the values given by Serrano et al. (1990), by Lange (1956), and by TRC (1998).

**Figure 1.** Variation of experimental (x) and literature values of viscosity, η , for pure *o*-xylene, against temperature, T : \square , Serrano et al., 1990; \triangle , Swain et al., 1997; \circ , Lange, 1956; ∇ , TRC, 1998.**Table 4. Coefficients a_j (mPa s) and Standard Deviations σ (mPa s) of Equation 1 for the γ -Butyrolactone + *o*-Xylene System**

	293.15 K	298.15 K	303.15 K	308.15 K
a_0	0.812	0.758	0.711	0.665
a_1	0.798	0.839	0.69	0.684
a_2	-0.15	-0.392	0.053	-0.117
a_3	0.698	0.919	0.159	0.266
a_4	-0.276	-0.38		
$\sigma \times 10^4$	0.5	3.8	5.8	7.2

Table 5. Coefficients a_j (mPa s) and Standard Deviations σ (mPa s) of Equation 1 for the γ -Butyrolactone + *m*-Xylene System

	293.15 K	298.15 K	303.15 K	308.15 K
a_0	0.618	0.588	0.555	0.527
a_1	0.715	0.555	0.461	0.38
a_2	0.067	0.366	0.686	0.589
a_3	0.482	0.102	-0.56	-0.344
a_4		0.134	0.471	0.343
$\sigma \times 10^4$	12	13.5	13.9	23

The experimental values of the viscosity were given as a power series by the equation

$$\eta = \sum_{j=0}^4 a_j x_1^j \quad (1)$$

where x_1 is the mole fraction of γ -butyrolactone. The obtained coefficients a_j and the deviations are presented in Tables 4 and 5. The experimental data and the smoothing curves are plotted in Figures 2 and 3.

Many equations trying to predict the viscosity of a mixture from the viscosities of the pure components and the composition have been proposed. Among them are the Hind-McLaughlin-Ubbelohde (1960) equation:

$$\eta = x_1^2 \eta_1 + 2x_1 x_2 \eta_{12} + x_2^2 \eta_2 \quad (2)$$

where η_i are the viscosities of the pure components, x_i are the mole fractions, η is the viscosity of the mixture, and η_{12} is a parameter dependent on unlike interactions.

The Heric (1966) equation, which for a binary system takes the form

$$\ln(\nu) = x_1 \ln(\nu_1 M_1) + x_2 \ln(\nu_2 M_2) - \ln(x_1 M_1 + x_2 M_2) + x_1 x_2 [\alpha_{12} + \alpha_{12}' (x_1 - x_2)] \quad (3)$$

In (3) ν_i and ν are the kinematic viscosities of the pure components and the mixture correspondingly, M_i is the molar mass of each component and α_{12} and α_{12}' are two adjustable parameters.

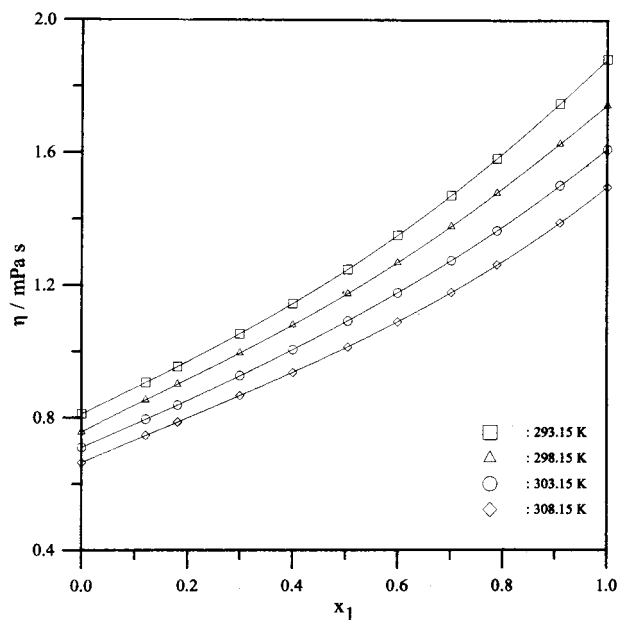


Figure 2. Variation of viscosity, η , with the mole fraction of γ -butyrolactone, x_1 , for the γ -butyrolactone + *o*-xylene system. Curves are least squares representation by eq 1.

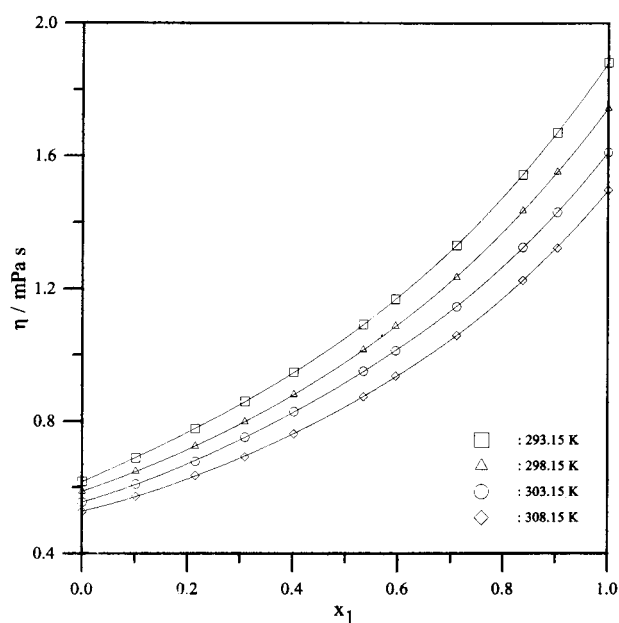


Figure 3. Variation of viscosity, η , with the mole fraction of γ -butyrolactone, x_1 , for the γ -butyrolactone + *m*-xylene system. Curves are least squares representations by eq 1.

The McAllister (1960) equation

$$\ln(\nu) = x_1^3 \ln(\nu_1) + 3x_1^2 x_2 \ln(\nu_{12}) + 3x_1 x_2^2 \ln(\nu_{21}) + x_2^3 \ln(\nu_2) - \ln(x_1 + x_2 M_2/M_1) + 3x_1^2 x_2 \ln[(2 + M_2/M_1)/3] + 3x_1 x_2^2 \ln[(1 + 2M_2/M_1)/3] + x_2^3 \ln(M_2/M_1) \quad (4)$$

in which ν_{12} and ν_{21} are also adjustable parameters, and the rest of the symbols have the meanings mentioned previously.

The experimental data were correlated to the above three equations, and the estimated parameters, as well as the average percentage deviations, σ^{perc} , defined by

Table 6. Calculated Parameters of Equations 2–4 and Average Percentage Deviation σ^{perc} for the γ -Butyrolactone + *o*-Xylene System

	293.15 K	298.15 K	303.15 K	308.15 K
eq 2				
$\eta_{12}/(\text{mPa s})$	1.149	1.103	1.019	0.942
$\sigma^{\text{perc}}_{\eta}(\text{eq 5a})$	0.47	0.57	0.52	0.95
eq 3				
α_{12}	0.130	0.187	0.165	0.153
α_{12}'	-0.024	-0.056	-0.052	-0.110
$\sigma^{\text{perc}}_{\nu}(\text{eq 5b})$	0.28	0.40	0.16	0.24
eq 4				
ν_{12}/cSt	1.391	1.302	1.223	1.113
ν_{21}/cSt	1.189	1.150	1.061	1.014
$\sigma^{\text{perc}}_{\nu}(\text{eq 5b})$	0.32	0.44	0.16	0.27

Table 7. Calculated Parameters of Equations 2–4 and Average Percentage Deviation σ^{perc} for the γ -Butyrolactone + *m*-Xylene System

	293.15 K	298.15 K	303.15 K	308.15 K
eq 2				
$\eta_{12}/(\text{mPa s})$	0.847	0.784	0.732	0.661
$\sigma^{\text{perc}}_{\eta}(\text{eq 5a})$	1.58	1.28	1.52	1.49
eq 3				
α_{12}	0.001	-0.047	-0.055	-0.132
α_{12}'	-0.040	0.019	-0.004	0.014
$\sigma^{\text{perc}}_{\nu}(\text{eq 5b})$	0.09	0.06	0.29	0.25
eq 4				
ν_{12}/cSt	1.229	1.163	1.088	1.003
ν_{21}/cSt	0.961	0.875	0.823	0.754
$\sigma^{\text{perc}}_{\nu}(\text{eq 5b})$	0.11	0.06	0.32	0.23

$$\sigma_{\eta}^{\text{perc}} = \frac{1}{n} \sum \frac{|\eta_{\text{exp}} - \eta_{\text{cal}}|}{\eta_{\text{exp}}} \times 100 \quad (5a)$$

$$\sigma_{\nu}^{\text{perc}} = \frac{1}{n} \sum \frac{|\nu_{\text{exp}} - \nu_{\text{cal}}|}{\nu_{\text{exp}}} \times 100 \quad (5b)$$

where n is the number of the experimental data, are presented in Tables 6 and 7. As one can see, in both systems and in the four temperatures, where measurements were made, eqs 3 and 4 correlate very well the experimental data and give about the same deviations, no more than 0.45% in any case. The same behavior of eqs 3 and 4 has been observed in other studied mixtures (Rodriguez et al., 1997). On the other hand, eq 2 gives larger deviations, lying in γ -butyrolactone + *o*-xylene system between 0.5 and 1% and in the γ -butyrolactone + *m*-xylene system at $\sim 1.5\%$.

Viscosity deviation, $\Delta\eta$, from linearity according to mole fraction was calculated from

$$\Delta\eta = \eta - x_1\eta_1 - x_2\eta_2 \quad (6)$$

Variation of $\Delta\eta$ with the mole fraction of γ -butyrolactone is presented in Figure 4 for 293.15 and 303.15 K, for both systems. Viscosity deviation is negative over the whole composition range for both mixtures and decreases in absolute value as the temperature is increased. For the same value of mole fraction, $\Delta\eta$ for the γ -butyrolactone + *m*-xylene system is almost 2-fold that of the γ -butyrolactone + *o*-xylene system.

Excess volumes, V^E , were calculated from density measurements, ρ , according to the equation

$$V^E = M_1 x_1 \left(\frac{1}{\rho} - \frac{1}{\rho_1} \right) + M_2 x_2 \left(\frac{1}{\rho} - \frac{1}{\rho_2} \right) \quad (7)$$

where ρ_i are the densities of the pure components and the

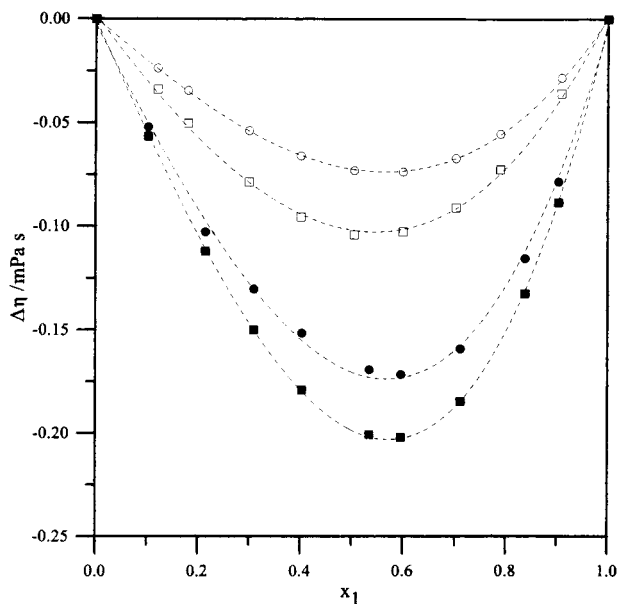


Figure 4. Dependence of viscosity deviation, $\Delta\eta$, on the mole fraction of γ -butyrolactone, x_1 : open symbols, γ -butyrolactone + *o*-xylene; full symbols, γ -butyrolactone + *m*-xylene, squares, 293.15 K; circles, 303.15 K.

Table 8. Coefficients b_j ($\text{cm}^3 \text{mol}^{-1}$) and Standard Deviations σ ($\text{cm}^3 \text{mol}^{-1}$) of Equation 8 for the γ -Butyrolactone + *o*-Xylene and γ -Butyrolactone + *m*-Xylene Systems

	288.15 K	293.15 K	298.15 K	303.15 K	308.15 K
<i>γ</i> -Butyrolactone + <i>o</i> -Xylene System					
b_0	-1.713	-1.785	-1.851	-1.896	-1.981
b_1	-0.526	-0.654	-0.575	-0.621	-0.49
b_2	0.775	0.93	0.679	0.409	0.329
b_3	-0.6				
σ	0.013	0.047	0.035	0.029	0.019
<i>γ</i> -Butyrolactone + <i>m</i> -Xylene System					
b_0	-2.089	-2.156	-2.227	-2.232	-2.289
b_1	-0.038	0.057	0.025	0.07	0.007
b_2	-0.236	-0.264	-0.291	-0.35	-0.39
b_3	-0.056	-0.031	-0.097		0.156
σ	0.016	0.016	0.014	0.01	0.017

rest of the symbols have the usual meaning. V^E values were correlated to the Redlich–Kister type equation (Redlich and Kister, 1948)

$$V^E = x_1 x_2 \sum_{j=0}^3 b_j (x_1 - x_2)^j \quad (8)$$

The values of coefficients b_j and the standard deviations are listed in Table 8. Dependence of excess volume on the mole fraction of γ -butyrolactone is presented in Figure 5 for the γ -butyrolactone + *o*-xylene system and in Figure 6 for the γ -butyrolactone + *m*-xylene system. By comparing the behavior of V^E for the two systems and by taking into account the corresponding figure for the γ -butyrolactone + *p*-xylene system (Avraam et al., 1998), one can see that for the three systems V^E is negative over the whole composition range and increases in absolute values as temperature increases. The largest deviations occur at $x_1 \sim 0.5$. Equimolar V^E , in absolute values, increases in the following sequence: *o*-xylene < *m*-xylene < *p*-xylene. The molar volumes of γ -butyrolactone, *o*-xylene, *m*-xylene, and *p*-xylene are, at 298.15 K, 76.6, 121.17, 123.42, and 123.94 $\text{cm}^3 \text{mol}^{-1}$, correspondingly. The molar volumes of the xylenes do not differ very much, but the different positions of the methyl groups changes the geometry of

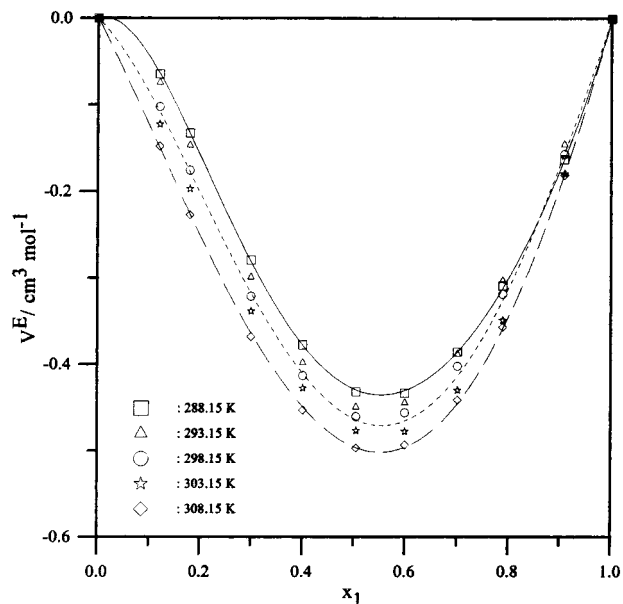


Figure 5. Variation of excess volume, V^E , with the mole fraction of γ -butyrolactone, x_1 , for the γ -butyrolactone + *o*-xylene system. Curves are least squares representations by eq 8 (—, 288.15 K; ···, 298.15 K; - - -, 308.15 K).

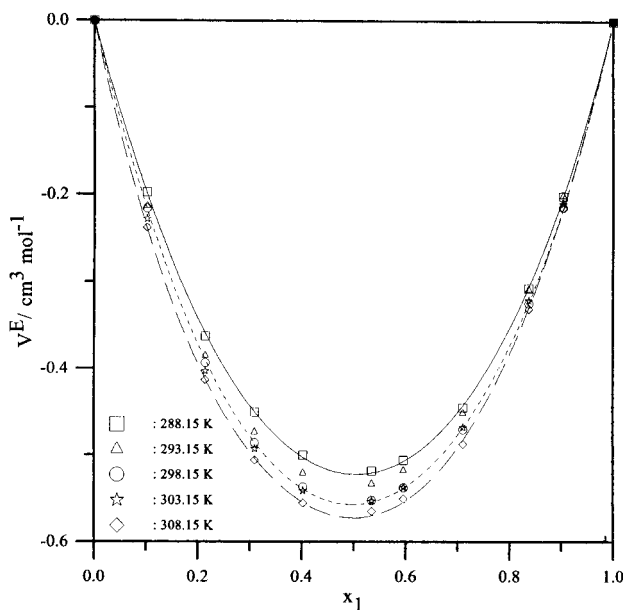


Figure 6. Variation of excess volume, V^E , with the mole fraction of γ -butyrolactone, x_1 , for the γ -butyrolactone + *m*-xylene system. Curves are least squares representations by eq 8 (—, 288.15 K; ···, 298.15 K; - - -, 308.15 K).

xylene molecules. The different size and the different geometry of the unlike molecules explains the negative V^E values, whereas dipole–dipole or dipole–induced dipole interactions between the unlike molecules could also be responsible (Palepu et al., 1987; Prigogine, 1957).

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