

# Excess Enthalpies of Binary Liquid Mixtures of $\gamma$ -Butyrolactone + Benzene, + Toluene, + Ethylbenzene, and + Carbon Tetrachloride, and Excess Volume of the $\gamma$ -Butyrolactone + Carbon Tetrachloride Liquid Mixture

B. S. Bjola,<sup>†</sup> M. A. Siddiqi,<sup>‡</sup> and P. Svejda\*

Ruhr-Universität Bochum, Institut für Thermo- und Fluidodynamik, D-44780 Bochum, Germany

Molar excess enthalpies of binary liquid mixtures of  $\gamma$ -butyrolactone + benzene, + toluene, + ethylbenzene, and + carbon tetrachloride were measured by means of a flow microcalorimeter of Picker type at 293.15 K. For the  $\gamma$ -butyrolactone + carbon tetrachloride mixture the molar excess volume was determined from density measurements at 293.15 K and 313.15 K. All measurements were performed at atmospheric pressure.

## Introduction

$\gamma$ -Butyrolactone is a biodegradable and water-soluble ester with a low degree of toxicity. It has applications in the chemical industry (as an intermediate in different syntheses, solvent, or catalyst) and power sources, electronics, specialty cleaning, foundry, textile, lithography, and coating industries.<sup>1,2</sup> However, despite its technological potential, thermodynamic data for  $\gamma$ -butyrolactone and its mixtures have been comparatively scarce. However, the measurements of various properties have been resumed in different laboratories, recently.<sup>3–7</sup>

For scientific reasons it is interesting to carry out thermodynamic studies of lactones and their mixtures. A comparison of their properties to those of the well-studied linear esters often reveals characteristic differences. Ring closure in  $\gamma$ -butyrolactone results in an exposure of the ester group (because of less steric screening), in an increasing polarizability (due to electron delocalization), and in a high ring strain, typical for small cycles.

This work continues the systematic investigation concerning the thermodynamic properties of lactone mixtures,<sup>4–7</sup> which will provide the necessary data for improving predictive group theories. In this paper, molar excess enthalpies of binary liquid mixtures of  $\gamma$ -butyrolactone + benzene, + toluene, + ethylbenzene, and + carbon tetrachloride at 293.15 K together with the molar excess volume of the  $\gamma$ -butyrolactone + carbon tetrachloride mixture at 293.15 K and 313.15 K are presented. All measurements were performed at atmospheric pressure.

## Experimental Section

$\gamma$ -Butyrolactone (oxolan-2-one,  $C_4H_6O_2$ ; Fluka A. G., Buchs, Switzerland), benzene (J. T. Baker Chem. B. V., Deventer, The Netherlands), toluene (J. T. Baker Chem.

B. V., Deventer, The Netherlands), ethylbenzene (Fluka A. G., Buchs, Switzerland), and carbon tetrachloride (J. T. Baker Chem. B. V., Deventer, The Netherlands) with stated purities of better than (99, 99, 99.5, 99, and 99) mol %, respectively, were stored over molecular sieves (0.4 nm, J. T. Baker Chem. B. V., Deventer, The Netherlands), which were activated at 573 K for more than 12 h under vacuum prior to use. The drying of the above-mentioned chemicals was performed with molecular sieves in the vapor phase, except for benzene and toluene, where it was carried out in the liquid phase. The purity of the chemicals was checked also by gas chromatography and was found to be in very good agreement with the original specifications, as described in a previous work.<sup>7</sup> All liquids were thoroughly degassed in a vacuum prior to use.

The measured densities and the calculated molar volumes of the lactone and the aromatics were the same as reported previously.<sup>4,5</sup> For carbon tetrachloride, densities of  $1.594\ 20\ g\cdot cm^{-3}$  at 293.15 K and  $1.555\ 22\ g\cdot cm^{-3}$  at 313.15 K were found, which may be compared with literature data of ( $1.594\ 20$ ,<sup>8</sup>  $1.594\ 17$ ,<sup>9</sup> and  $1.594\ 10$ <sup>10</sup>)  $g\cdot cm^{-3}$  at 293.15 K or ( $1.555\ 22$ ,<sup>8</sup>  $1.555\ 62$ ,<sup>9</sup> and  $1.555\ 15$ <sup>11</sup>)  $g\cdot cm^{-3}$  at 313.15 K.

Relative atomic masses,<sup>12</sup>  $A_r(H) = 1.007\ 95$ ,  $A_r(C) = 12.011$ ,  $A_r(O) = 15.9994$ , and  $A_r(Cl) = 35.4528$ , were used for the calculation of the molar volumes from densities.

The molar excess volumes  $V^E$  were calculated from the precise densities of the liquids which were measured by means of a vibrating glass tube densimeter (DMA 02 D, Chempro Paar, Germany) at 293.15 K and 313.15 K and atmospheric pressure. The preparation of the samples (from carefully degassed liquids, under vacuum, with vapor space correction), their transfer to the vibrating tube (without losses by evaporation or contact to air), and the experimental setup (like measurement and control of the temperature) have been described in a previous paper.<sup>13</sup> In summary, the following uncertainties were found:  $\pm 0.003$  K for the temperature;  $3 \times 10^{-5}$  for the final mole fractions;  $2 \times 10^{-5}\ g\cdot cm^{-3}$  at 293.15 K and  $1 \times 10^{-4}\ g\cdot cm^{-3}$  at 313.15 K for the density, with a reproducibility of  $3 \times 10^{-6}\ g\cdot cm^{-3}$ ; and better than  $0.003\ cm^3\cdot mol^{-1}$  for the excess molar volume at about equimolar composition.

\* Corresponding author. Fax: +49-234 32 14280. E-mail: peter.svejda@ruhr-uni-bochum.de.

<sup>†</sup> Permanent address: Politehnica University of Bucharest, Dept. Applied Physical Chemistry, Str. Polizu nr. 1, Ro-78126 Bucharest, Romania.

<sup>‡</sup> Gerhard Mercator Universität Duisburg, Fachbereich 7, Thermodynamik, Lotharstr. 1, D-47057 Duisburg, Germany.

**Table 1. Density  $\rho$  and Molar Excess Volume  $V^E$  of the Binary Liquid Mixture of  $\gamma$ -Butyrolactone (1) + Carbon Tetrachloride (2) at Temperatures of 293.15 K and 313.15 K and Atmospheric Pressure**

$x_1$	$\rho/\text{g}\cdot\text{cm}^{-3}$		$V^E/\text{cm}^3\cdot\text{mol}^{-1}$	
	293.15 K	313.15 K	293.15 K	313.15 K
0.000 00	1.594 20	1.555 22		
0.104 58	1.559 68	1.522 63	-0.289	-0.307
0.202 13	1.524 75	1.489 73	-0.491	-0.535
0.301 36	1.486 54	1.453 64	-0.641	-0.709
0.406 90	1.443 28	1.412 36	-0.767	-0.839
0.451 79	1.423 76	1.393 62	-0.794	-0.863
0.506 50	1.399 20	1.370 16	-0.815	-0.888
0.604 61	1.352 48	1.325 34	-0.791	-0.861
0.698 79	1.304 47	1.279 12	-0.703	-0.765
0.801 47	1.248 48	1.225 03	-0.534	-0.578
0.906 93	1.186 88	1.165 39	-0.278	-0.302
1.000 00	1.129 20	1.109 40		

The molar excess enthalpies  $H^E$  were measured with a dynamic flow calorimeter of Picker type (Setaram, France) at 293.15 K and at atmospheric pressure, as has been described in detail elsewhere.<sup>14</sup> The composition of each mixture was determined by its density, which was measured in a thermostatically controlled flow densimeter

(DMA 602, Chempro Paar, Germany, vibrating glass tube model). Then, from these measurements and by means of previous Redlich–Kister correlations of excess volume versus concentration at 293.15 K,<sup>4,5,7</sup> the mole fraction could be calculated with an estimated uncertainty of  $\pm 0.0001$ . Since no excess volume data of the  $\gamma$ -butyrolactone + carbon tetrachloride liquid mixture were available in the literature, these have been measured during this work too and are reported here. The calorimeter was thermostated to  $\pm 0.03$  K or better and controlled by calibrated Pt-100 thermoresistors. It was checked against literature data of the benzene + cyclohexane system<sup>15</sup> and exhibited an uncertainty of less than 1% at the maximum value of  $H^E$ .

## Results

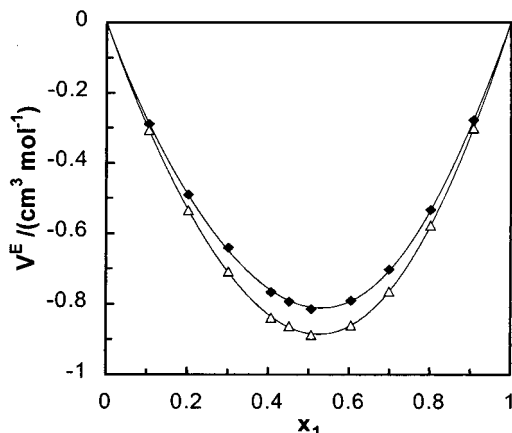
The experimental values of the molar excess volume  $V^E$  and molar excess enthalpies  $H^E$  are summarized in Tables 1 and 2, respectively, at various mole fractions  $x_i$  of the mixtures. All excess properties have been fitted to Redlich–

**Table 2. Molar Excess Enthalpies  $H^E$  of Binary Liquid Mixtures with  $\gamma$ -Butyrolactone as One Component at 293.15 K and Atmospheric Pressure**

$x_1$	$H^E/\text{J}\cdot\text{mol}^{-1}$	$x_1$	$H^E/\text{J}\cdot\text{mol}^{-1}$	$x_1$	$H^E/\text{J}\cdot\text{mol}^{-1}$	$x_1$	$H^E/\text{J}\cdot\text{mol}^{-1}$
$\gamma$ -Butyrolactone (1) + Benzene (2)							
0.0036	4.6	0.2073	3.0	0.4453	-0.8	0.7885	26.8
0.0080	7.8	0.2425	-0.8	0.4870	1.6	0.7911	27.4
0.0145	13.0	0.2600	-1.5	0.5195	4.6	0.7942	28.5
0.0265	19.1	0.2786	-3.7	0.5482	7.8	0.8140	27.3
0.0504	23.8	0.2941	-4.1	0.5936	11.8	0.8253	27.3
0.0592	24.0	0.3151	-4.3	0.5985	13.6	0.8458	26.6
0.0691	23.9	0.3259	-4.7	0.6463	18.1	0.8537	25.4
0.0723	23.6	0.3334	-4.3	0.6524	19.6	0.8677	24.9
0.0757	23.5	0.3591	-5.3	0.6928	23.4	0.8743	24.5
0.1053	18.0	0.3700	-3.5	0.6984	23.3	0.8803	23.1
0.1075	19.5	0.3778	-4.7	0.7258	25.3	0.9080	20.6
0.1141	18.3	0.3796	-2.9	0.7314	25.5	0.9226	17.8
0.1460	11.1	0.3983	-2.6	0.7583	26.8	0.9465	13.7
0.1818	5.4	0.4159	-3.0	0.7613	26.9	0.9667	8.8
$\gamma$ -Butyrolactone (1) + Toluene (2)							
0.0082	17.9	0.3735	202.8	0.6403	186.0	0.8616	101.4
0.0409	66.4	0.3986	207.8	0.6555	184.5	0.8734	94.0
0.1308	137.4	0.4440	205.8	0.6976	172.7	0.9026	73.5
0.1547	151.1	0.4631	209.1	0.7143	167.2	0.9211	62.1
0.2218	174.3	0.5102	204.1	0.7544	151.9	0.9439	45.3
0.2413	181.9	0.5212	205.9	0.7627	148.9	0.9651	29.2
0.3005	192.9	0.5793	197.5	0.8060	130.9	0.9894	8.3
0.3209	198.6	0.5835	198.2	0.8193	124.5		
$\gamma$ -Butyrolactone (1) + Ethylbenzene (2)							
0.0169	43.7	0.2658	339.4	0.5933	401.3	0.8390	236.0
0.0257	56.9	0.3087	366.2	0.6174	394.3	0.8698	202.7
0.0428	97.7	0.3467	383.8	0.6504	381.3	0.8919	171.5
0.0708	141.4	0.3887	400.4	0.6766	368.2	0.9062	154.0
0.0986	185.7	0.4216	404.6	0.7088	350.5	0.9215	130.2
0.1187	213.9	0.4554	411.6	0.7347	330.9	0.9383	102.7
0.1390	234.0	0.4940	413.3	0.7621	310.4	0.9509	85.1
0.1773	272.7	0.5257	412.7	0.7887	286.5	0.9708	49.7
0.2199	309.9	0.5618	407.2	0.8172	260.0	0.9827	31.7
$\gamma$ -Butyrolactone (1) + Carbon Tetrachloride (2)							
0.0026	15.0	0.2552	322.3	0.7193	74.0	0.9588	1.1
0.0028	18.0	0.3046	316.5	0.7258	73.6	0.9620	0.8
0.0113	62.6	0.3676	296.1	0.7765	45.4	0.9708	-0.2
0.0170	85.6	0.3724	291.6	0.7951	38.3	0.9727	0.0
0.0228	110.7	0.4387	258.2	0.8093	35.0	0.9741	-0.3
0.0253	120.1	0.4881	227.5	0.8380	25.1	0.9772	-0.8
0.0355	148.5	0.5312	199.5	0.8400	22.6	0.9792	-1.6
0.0372	156.0	0.5734	170.2	0.8707	13.0	0.9835	-1.9
0.0458	179.5	0.5996	153.6	0.8767	10.8	0.9844	-2.8
0.0523	189.9	0.6063	150.1	0.9083	6.5	0.9863	-3.8
0.0697	222.2	0.6467	121.2	0.9206	3.5	0.9882	-4.0
0.0799	238.8	0.6861	96.4	0.9432	2.0	0.9915	-2.2
0.1159	278.1	0.6886	98.6	0.9447	1.9	0.9921	-2.4
0.1662	307.2	0.6940	93.3	0.9451	1.6		
0.2005	318.2	0.7144	78.0				

**Table 3. Coefficients  $A_i$  and Standard ( $\sigma$ ) and Maximum ( $\delta$ ) Deviations of the Redlich–Kister Equation (Eq 1) for the Experimental Excess Data at Temperature  $T$  (for  $V^E$  All  $A_i$ ,  $\sigma$ , and  $\delta$  in the Units  $\text{cm}^3\cdot\text{mol}^{-1}$ ; for  $H^E$  All  $A_i$ ,  $\sigma$ , and  $\delta$  in the Units  $\text{J}\cdot\text{mol}^{-1}$ )**

excess property	$T/\text{K}$	$A_0$	$A_1$	$A_2$	$A_3$	$A_4$	$A_5$	$A_6$	$A_7$	$\sigma$	$\delta$
$\gamma$ -Butyrolactone (1) + Benzene (2)											
$H^E$	293.15	11.3	181.0	255.8	-130.1	-314.8	164.3	704.8	-595.2	0.6	1.2
$\gamma$ -Butyrolactone (1) + Toluene (2)											
$H^E$	293.15	824.5	-123.8	278.4	24.7	-6.2	-490.2	365.6		1.3	2.5
$\gamma$ -Butyrolactone (1) + Ethylbenzene (2)											
$H^E$	293.15	1656.6	-17.8	249.8	-85.1	286.2	-271.4			1.6	3.5
$\gamma$ -Butyrolactone (1) + Carbon Tetrachloride (2)											
$H^E$	293.15	876.2	-1270.2	669.1	-884.0	-897.8	2006.0	2180.1	-2863.3	1.8	3.4
$V^E$	293.15	-3.234	-0.414	0.078	0.431					0.005	0.008
$V^E$	313.15	-3.535	-0.367	0.169	0.275					0.003	0.004

**Figure 1.** Molar excess volume  $V^E$  versus mole fraction  $x_1$  of the binary liquid mixture of  $\gamma$ -butyrolactone (1) + carbon tetrachloride (2) at 293.15 K ( $\blacklozenge$ ) and 313.15 K ( $\triangle$ ) and atmospheric pressure.

Kister type polynomials

$$Z_{\text{calc}}^E = x_1 x_2 \left[ \sum_{i=0}^k A_i (x_1 - x_2)^i \right] \quad (1)$$

where  $Z^E = V^E/\text{cm}^3\cdot\text{mol}^{-1}$  or  $H^E/\text{J}\cdot\text{mol}^{-1}$ .

The coefficients  $A_i$  are summarized in Table 3 together with the standard deviation  $\sigma$ , defined as

$$\sigma^2 = \sum_N [Z^E - Z_{\text{calc}}^E]^2 / (N - m) \quad (2)$$

where  $Z^E$  is the experimental excess property,  $N$  is the number of data points, and  $m$  is the number of coefficients  $A_i$ , and the maximum deviation  $\delta$

$$\delta = \max |Z^E - Z_{\text{calc}}^E| \quad (3)$$

The coefficients  $A_i$  and the deviations  $\sigma$  and  $\delta$  are given in the same units as the corresponding excess properties.

It was found that the measured data of the peculiarly shaped  $H^E$  versus  $x$  curve for  $\gamma$ -butyrolactone + benzene or + carbon tetrachloride could be fitted much better by Redlich–Kister polynomials (eq 1) using many coefficients than by rational functions, which are sometimes used in the literature for skewed excess curves too.

## Discussion

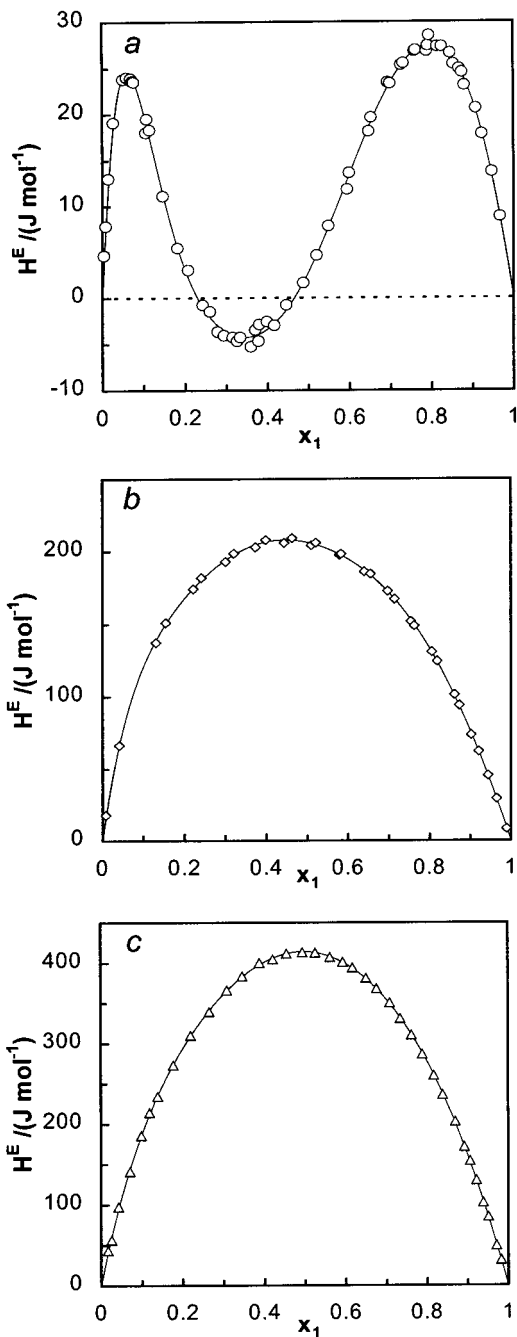
For the molar excess volume of the mixture of  $\gamma$ -butyrolactone + carbon tetrachloride, only negative values have been found, both at 293.15 K and 313.15 K. The corresponding Redlich–Kister plots (Figure 1) are almost symmetrical, with minima of about  $-0.82 \text{ cm}^3\cdot\text{mol}^{-1}$  and  $-0.89$

$\text{cm}^3\cdot\text{mol}^{-1}$ , respectively. These curves are quite similar to previously reported  $V^E$  functions for binary systems of  $\gamma$ -butyrolactone with different aromatic solvents,<sup>4,5,16</sup> namely that all are rather symmetrical and that the minima are always deeper at the higher temperature. However, the mixture with  $\text{CCl}_4$  shows deeper minima than the systems with aromatics.

The molar excess enthalpies of the binary systems containing  $\gamma$ -butyrolactone + benzene, or + toluene, or + ethylbenzene are plotted in Figure 2 and exhibit significant differences in shape and maximum value. The most interesting results were obtained for the mixture of  $\gamma$ -butyrolactone + benzene, where a pronounced M-shaped curve, with two maxima and one minimum, has been observed. (Note in Figure 2a that a large number of experimental points has been measured to verify the shape of the curve as well as possible.) In this system, all the measured excess enthalpies are lying in a relatively narrow range of less than  $35 \text{ J}\cdot\text{mol}^{-1}$ . That means the mixture is almost athermal, which allows us to observe small heat variations due to changes of the liquid structure during the mixing process. In comparison, the binary mixtures of  $\gamma$ -butyrolactone with toluene and ethylbenzene exhibit quite usual  $H^E$  curves with positive values and maxima of about  $200 \text{ J}\cdot\text{mol}^{-1}$  and  $420 \text{ J}\cdot\text{mol}^{-1}$ , respectively.

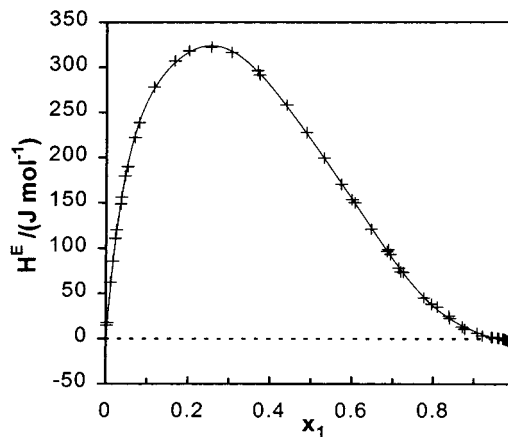
Endothermic mixing processes are common in systems consisting of a polar + a nonpolar liquid.<sup>14,17</sup> Structural order which exists in pure polar liquids is disturbed or broken up during mixing, which consumes energy and leads to the observed endothermic effect. Thus, in the pure liquid state the lactone will develop a fairly strong molecular order, due to its large dipole moment.<sup>7</sup> Similarly, a weaker order is found for pure aromatics as a consequence of the polarizable  $\pi$ -system. By mixing, these orders are destroyed and endothermic  $H^E$  values are observed, like in the systems of lactone + toluene or + ethylbenzene. However, the dipole of the lactone may also strongly attract the  $\pi$ -system of the aromatics<sup>18</sup> (dipole-induced dipole interactions<sup>19</sup>), leading to an exothermic effect. And indeed, in the system  $\gamma$ -butyrolactone + benzene apparently the shape and the size of both molecules and their mutual interactions are just right to attract each other in an optimal way in their mixture at mole fractions of the lactone around 0.3. The observed M-shaped  $H^E$  curve results from a superposition of this exothermic effect on the usual endothermic curve. A  $\gamma$ -butyrolactone mole fraction of 0.3 could mean that at the minimum of  $H^E$  on the average two benzene molecules are "coordinated" tightly with one lactone molecule.

The system of  $\gamma$ -butyrolactone + carbon tetrachloride is related to that of the lactone + benzene. Its excess enthalpy exhibits an asymmetrical S-shaped curve, mainly positive and not exceeding  $330 \text{ J}\cdot\text{mol}^{-1}$  (Figure 3). Slight, but

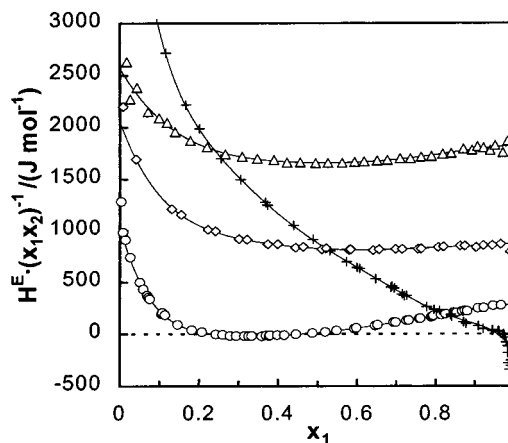


**Figure 2.** Molar excess enthalpy  $H^E$  versus mole fraction  $x_1$  of the binary liquid mixtures of  $\gamma$ -butyrolactone (1): (a) + benzene (2), (b) + toluene (2), or (c) + ethylbenzene (2) at 293.15 K and atmospheric pressure.

reproducible, exothermal effects (with a minimum value of about  $-5 \text{ J mol}^{-1}$ , cf. Table 2) were found for a narrow composition range at high lactone concentrations, again after very careful measurements. In this mixture there exists also a strong attraction between the lactone and carbon tetrachloride, possibly due to the interaction of the dipole of the lactone with the strongly polarizable C-Cl bonds. Superposition of the corresponding exothermal effect on the normal endothermal curve this time results in the S-shaped  $H^E$  curve. However, the position of the minimum of  $H^E$  at large mole fractions of lactone seems to indicate that several lactone molecules interact strongly with one carbon tetrachloride molecule.  $H^E$  curves of a similar shape have been reported for other binary mixtures of carbon tetrachloride + lower aliphatic alcohols.<sup>20</sup>



**Figure 3.** Molar excess enthalpy  $H^E$  versus mole fraction  $x_1$  of the binary liquid mixture of  $\gamma$ -butyrolactone (1) + carbon tetrachloride (2) at 293.15 K and atmospheric pressure.



**Figure 4.** Reduced molar excess enthalpy  $[H^E/(x_1 x_2)]$  versus mole fraction  $x_1$  of the binary liquid mixtures of  $\gamma$ -butyrolactone (1): (○) + benzene (2), (◇) + toluene (2), (△) + ethylbenzene (2), or (+) + carbon tetrachloride (2) at 293.15 K and atmospheric pressure.

It seems of interest that plots of the reduced molar excess enthalpy  $[H^E/(x_1 x_2)]$  versus mole fraction  $x_1$  of the measured data (Figure 4) support these interpretations nicely. Such plots showing partial molar enthalpies are especially sensitive to the shape of a curve of excess properties. Previously, it was found that formation of a strong complex in a liquid mixture would exhibit a sharp minimum in the graph.<sup>21</sup> In the present mixtures of  $\gamma$ -butyrolactone, which is much less polar, only a very broad and shallow minimum can be observed in the mixture with benzene again at a mole fraction of about 0.3. At higher values of  $x_1$  the curve becomes almost a straight line, suggesting rather an ideal behavior of the mixture, as discussed above. The minimum shifts to  $x_1 \approx 0.5$  in the mixtures of  $\gamma$ -butyrolactone + toluene or + ethylbenzene, because in the looser structure of these two benzene derivatives the mixing partners seem to be coordinated only 1:1. The curve of  $[H^E/(x_1 x_2)]$  for  $\gamma$ -butyrolactone + carbon tetrachloride reflects a completely different behavior at mixing compared to that of the aromatics. There is no minimum at low mole fractions, no ideal mixing is shown, but a surprisingly sharp minimum is found at very high values of  $x_1$ , where a single carbon tetrachloride molecule seems to be coordinated very strongly with many lactone particles.

#### Acknowledgment

Useful comments by the reviewers were very much appreciated.



## Literature Cited

- (1) *Ullmann's Encyclopedia of Industrial Chemistry*, 6th ed.; Wiley-VCH Weinheim: Berlin, 2000.
- (2) Cummins, D.; Boschloo, G.; Ryan, M.; Corr, D.; Rao, S. N.; Fitzmaurice, D. Ultrafast Electrochromic Windows Based on Redox-Chromophore Modified Nanostructured Semiconducting and Conducting Films. *J. Phys. Chem. B* **2000**, *104*, 11449–11459.
- (3) Ritzoulis, G.; Missopolinou, D.; Doulami, S.; Panayiotou, C. Relative Permittivities, Densities, Refractive Indices and Ultrasound Velocities of the Binary Systems of  $\gamma$ -Butyrolactone with Methanol, Ethanol, 1-Butanol, and 1-Octanol. *J. Chem. Eng. Data* **2000**, *45*, 636–641.
- (4) Klein, A.; Svejda, P. Isothermal Vapor-Liquid Equilibria and Excess Volumes of Binary Mixtures of Benzene +  $\gamma$ -Butyrolactone,  $\gamma$ -Valerolactone,  $\delta$ -Valerolactone, or  $\epsilon$ -Caprolactone at 293.15K and 313.15K. *ELDATA Int. Electron. J. Phys. Chem. Data* **1995**, *1*, 87–94.
- (5) Fornefeld-Schwarz, U.; Svejda, P. Excess Volumes of Liquid Mixtures of  $\gamma$ -Butyrolactone (Oxalan-2-one),  $\gamma$ -Valerolactone (5-Methylloxalan-2-one),  $\delta$ -Valerolactone (Oxan-2-one), or  $\epsilon$ -Caprolactone (Oxepan-2-one) + Toluene or + Ethylbenzene at 293.15K and 313.15K. *ELDATA Int. Electron. J. Phys. Chem. Data* **1997**, *3*, 17–24.
- (6) Fornefeld-Schwarz, U.; Klein, A.; Svejda, P. Densities and Isothermal Compressibilities of Liquid Mixtures of  $\gamma$ -Butyrolactone,  $\gamma$ -Valerolactone,  $\delta$ -Valerolactone, or  $\epsilon$ -Caprolactone + Benzene, + Toluene, or Ethylbenzene at 293.15K and Pressures up to 10 MPa. *ELDATA Int. Electron. J. Phys. Chem. Data* **1997**, *3*, 131–148.
- (7) Fornefeld-Schwarz, U. M.; Svejda, P. Refractive Indices and Relative Permittivities of Liquid Mixtures of  $\gamma$ -Butyrolactone,  $\gamma$ -Valerolactone,  $\delta$ -Valerolactone, or  $\epsilon$ -Caprolactone + Benzene, + Toluene, or + Ethylbenzene at 293.15K and 313.15K and Atmospheric Pressure. *J. Chem. Eng. Data* **1999**, *44*, 597–604.
- (8) Siddiqi, M. A.; Götze, G.; Kohler, F. Excess Volumes of Mixtures of Cycloalkanes and Methylcycloalkanes. *Ber. Bunsen-Ges. Phys. Chem.* **1980**, *44*, 529–536.
- (9) TRC Tables 23-10-2-(10.011)-a, -d. C-Cl-H. Chloroalkanes, C<sub>1</sub> and C<sub>2</sub>. *TRC Thermodynamic Tables—Non Hydrocarbons*; Thermodynamics Research Center: Texas A&M University, College Station, TX, pp a-7240, d-7240 (edition up to December 31, 1999).
- (10) Boublik, T.; Aim, K. Heats of Vaporisation of Simple Non-Spherical Molecule Compounds. *Collect. Czech. Chem. Commun.* **1972**, *37*, 3513–3521.
- (11) Subach, D. J.; Kong, C. L. Thermodynamics of Solutions: Excess Volumes of Benzene, Carbon Tetrachloride, and Mesitylene Mixtures. *J. Chem. Eng. Data* **1973**, *18*, 403–405.
- (12) IUPAC Commission on Atomic Weights and Isotopic Abundances. Atomic Weights of the Elements 1993. *J. Phys. Chem. Ref. Data* **1995**, *24*, 1561–1575.
- (13) Svejda, P.; Siddiqi, M. A.; Hahn, G.; Cristoph, N. Excess Volume, Isothermal Compressibility and Excess Enthalpy of the Binary Liquid System Mixtures 2,2,2-Trifluoroethanol + 2,5,8,11,14-Pentaoxapentadecane. *J. Chem. Eng. Data* **1990**, *35*, 47–49.
- (14) Hahn, G.; Svejda, P.; Kehiaian, H. V. Excess Enthalpies of the Liquid Systems: 1,2-Dichloroethane + *n*-Alkanes or + 2,2,4-Trimethylpentane. *Fluid Phase Equilib.* **1986**, *28*, 309–323.
- (15) Elliot, K.; Wormald, C. J. A Precision Differential Flow Calorimeter. The Excess Enthalpy of Benzene + Cyclohexane Between 280.15 K and 393.15 K. *J. Chem. Thermodyn.* **1976**, *8*, 881–893.
- (16) Avraam, T.; Moumouzias, G.; Ritzoulis, G. A Study on Excess Volumes and Dielectric Properties in the  $\gamma$ -Butyrolactone + *p*-Xylene System at Various Temperatures. *J. Chem. Eng. Data* **1998**, *43*, 51–54.
- (17) Hahn, G.; Svejda, P. Excess Volumes and Excess Enthalpies of Binary Liquid Mixtures of *trans*-1,2-Dichloroethene and of *cis*-1,2-Dichloroethene + *n*-Alkanes or + 2,2,4-Trimethylpentane. *J. Chem. Eng. Data* **1996**, *41*, 619–623.
- (18) Luhmer, M.; Bartik, K.; Dejaegere, A.; Bovy, P.; Reisse, J. The Importance of Quadrupolar Interactions in Molecular Recognition Processes Involving a Phenyl Group. *Bull. Soc. Chim. Fr.* **1994**, *131*, 603–606.
- (19) Israelachvili, J. N. *Intermolecular & Surface Forces*, 2nd ed.; Academic Press: London, 1992; Chapter 5.
- (20) Otterstedt, J.-E. A.; Missen, R. W. Thermodynamic Properties of Solutions of Alcohols and Carbon Tetrachloride. *Trans. Faraday Soc.* **1962**, *58*, 879–889.
- (21) Munn, R. J.; Svejda, P. Excess Enthalpy of Mixing of Propionic Acid + Triethylamine by Flow-Calorimetric Determination of Difference Enthalpies of Mixing. *Ber. Bunsen-Ges. Phys. Chem.* **1979**, *83*, 920–925.

Received for review March 18, 2001. Accepted May 16, 2001.

JE010091V