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Isothermal Vapor−Liquid Equilibria Measurements, Excess Molar Enthalpies, and Excess Molar Volumes of Dimethyl Carbonate + Methanol, + Ethanol, and + Propan-1-ol at 313.15 K

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Isothermal vapor−liquid equilibria (VLE), excess molar enthalpies, H_m^E, and excess molar volumes, V_m^E, have been measured for the binary mixtures dimethyl carbonate + methanol, + ethanol, or + propan-1-ol at 313.15 K and at atmospheric pressure. The three mixtures exhibit large deviations from ideality, showing an azeotrope at 0.14, 0.42, and 0.78 mole fraction of dimethyl carbonate, respectively. The VLE data were correlated by the Wilson equation and the appropriate parameters are reported. H_m^E and V_m^E values have been correlated using the Redlich−Kister equation by the method of least-squares.

Introduction

The thermodynamic study of esters of carbonic acid is of considerable interest owing to their uses in industry for the synthesis of many chemicals, in pharmaceuticals, and in agricultural chemistry; such chemicals are also used as extractive solvents.

Continuing our research program on the thermodynamic properties of binary mixtures containing dialkyl carbonates (Francesconi and Comelli, 1995, 1996; Comelli et al., 1996), we report in this paper isothermal vapor−liquid equilibria (VLE) measurements, excess molar enthalpies, H_m^E, and excess molar volumes, V_m^E, of the binary mixtures dimethyl carbonate + methanol, + ethanol, and + propan-1-ol at 313.15 K and at atmospheric pressure.

We have not found in literature any data on the thermodynamic properties of the three binary mixtures studied in this paper.

Experimental Section

Chemicals. Dimethyl carbonate, an Aldrich product, stated purity 99 mol %, was fractionally distilled with sodium, as described in a previous paper (Francesconi and Comelli, 1996) and the purity, tested by GCL after purification, was 99.9 mol %.

Methanol and propan-1-ol, Aldrich products with stated purity +99.9 and 99.7 mol %, and ethanol, a Fluka product, puriss., purity > 99.8 mol %, were used without further purification since no appreciable impurities were detected by GLC analysis.

Before use, all chemicals were kept in dark bottles, stored over molecular sieves (Union Carbide $^1/_{16}$ in. rods, pore diameter 4A) and degassed using an ultrasonic bath (Hellma, type 460, Milan, Italy).

Table 1 reports density values at (298.15 and 313.15) K and boiling points in comparison with literature values.

Apparatus and Procedure. Isothermal VLE measurements were determined using a glass dynamic recirculating still (Fritz Gmbh, Normag, Hofheim, Germany), the same as used in our previous studies (Francesconi and Comelli, 1996; Comelli et al. 1996). The experimental method and the operating procedure have been described elsewere (Gmehling et al., 1980).

Pressure and temperature were determined to a precision of ±0.05 kPa and ±0.05 K by digital instruments (Fritz Gmbh, Normag) supplied with the still.

In each measurement, the temperature was fixed at 313.15 K and the liquid mixture was kept at the boiling point for 20 min to reach the stationary state. Then samples of ≈1.5 cm^3 of liquid and condensed vapor from the Cottrel pump were taken and analyzed by a digital density meter (Anton Paar, model DMA 60, Graz, Austria) equipped with a measuring cell (Anton Paar, type DMA 602), with a precision of the measured oscillation period τ of 1×10^{-6} s.

The apparatus and the operating procedure have been described elsewere (Fermeglia and Lapasin, 1988).

The temperature of the U-shaped tube was checked using a digital thermometer (Anton Paar, DT-40) with an accuracy of ±0.01 K, while an external Hethotherm bath circulator (Heto type 01 DBT 623, Birkerød, Denmark) maintained the temperature constant to within ±0.005 K. Densities, ρ, were obtained at (313.15 ± 0.01) K with a precision of 1×10^{-5}g·cm^{-3}. The apparatus was calibrated with dry air (Kohlrausch, 1968) and bidistilled water (Wagenbreth and Blanke, 1971); we have adopted a period of $\tau = 10\,000$ units for density determinations.

Calibration curves, necessary to determine mole fraction of the samples, were determined by measuring the densities of the solutions prepared by mass, weighing each component with a precision balance (Mettler, model AE 160, precision ±0.0001 g) and following the procedure described elsewere (Fermeglia and Lapasin, 1988).

The reported mole fractions of dimethyl carbonate are precise to ±0.002 for VLE measurements.

We have used the same density meter also to determine the excess molar volumes, V_m^E, of the three mixtures. Before measurements, the apparatus was checked with the test mixture benzene + cyclohexane (Wilhelm, 1985): the agreement between the measured and the literature values was better than 0.5% for the excess molar volume, V_m^E, in the range $0.1 < x < 0.9$ of the mole fraction of benzene.

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Table 1. Densities, ρ, and Boiling Points, T_b, of Pure Components and Comparison with Literature Values

	ρ(298.15K)/g·cm^{-3}		ρ(313.15K)/g·cm^{-3}		T_b(101.3kPa)/K	
component	exptl	lit.	exptl	lit.	exptl	lit.
dimethyl carbonate	1.063 28	1.063 50[a] 1.063 059[c]	1.043 41		363.60	363.65[b]
methanol	0.786 56	0.786 37[d] 0.786 6[f] 0.786 8[g]	0.772 29	0.7723[e]	337.70	337.696[d]
ethanol	0.785 09	0.784 93[d] 0.785 0[e] 0.785 4[g]	0.772 22	0.7721[e]	351.45	351.443[d]
propan-1-ol	0.799 58	0.799 60[d] 0.799 5[e] 0.799 2[g]	0.787 29	0.7873[e]	370.30	370.301[d]

[a] Garcia de la Fuente et al., 1992. [b] CRC Handbook, 1967. [c] Negadi et al., 1993. [d] Riddick et al., 1986. [e] Arce et al., 1995. [f] Nikam et al., 1995.

Table 2. Comparison of Vapor Pressure $P°_i$/kPa of Pure Components at 313.15 K with Literature Data

	$P°$/kPa	
component	present paper	lit.
dimethyl carbonate	15.05	14.92[a]
methanol	35.45	35.428[b] 35.484[c] 35.429[d]
ethanol	17.90	17.877[b] 17.910[e] 17.909[f]
propan-1-ol	6.90	6.924[b] 6.921[g]

[a] Negadi et al., 1993. [b] Riddick et al., 1986. [c] Toghiani et al., 1994. [d] Hiaki et al., 1994. [e] Kurihara et al., 1993. [f] Vercher et al., 1996. [g] Hiaki et al., 1995.

Solutions were prepared by mass in ≈15 cm^3 capacity flasks, and corrections for buoyancy and evaporation of the components were made.

The excess molar volume, V_m^E, has been calculated from density measurements using the following equation

$$V_m^E/(\text{cm}^3\cdot\text{mol}^{-1}) = (x_1M_1 + x_2M_2)/\rho - x_1M_1/\rho_1 - x_2M_2/\rho_2 \tag{1}$$

where x_i, ρ_i, ρ, and M_i represent the mole fraction, the density of pure component and of mixture, and the molar mass, respectively.

The accuracies of V_m^E values are less than 0.002 cm^3·mol^{-1}.

An LKB flow microcalorimeter (model 2107, LKB produkter, Bromma, Sweden) was used to determine the excess molar enthalpies, H_m^E, of the mixtures. The apparatus, the electrical calibration, and its operating procedure have been described elsewere (Monk and Wadso, 1968; Francesconi and Comelli, 1986). Two automatic burets (ABU, Radiometer, Copenhagen, Denmark) were used to pump liquids through the mixing cell of the calorimeter. The temperature was kept constant at (313.15 ± 0.01) K. The performance of the calorimeter was checked by measuring the H_m^E of the test mixture hexane + cyclohexane (Gmehling, 1993). Agreement with literature is better than 0.5% over the central range of mole fraction of hexane.

The uncertainties in mole fraction and H_m^E are estimated to be 0.0005 and 0.5%, respectively.

Results and Discussion

The experimental vapor pressures of pure components, $P°_i$, which are necessary in the analysis of the VLE data, were determined using the same circulating still used for mixtures and are listed in Table 2 in comparison with literature data.

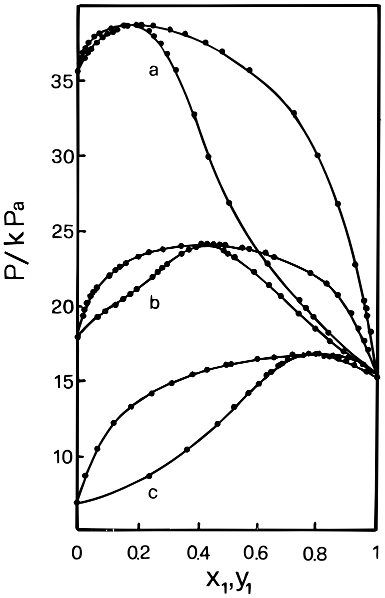

Figure 1. $P-x_1-y_1$ plots for the dimethyl carbonate + 1-alkanol (2) mixtures at 313.15 K. (●) Experimental points; solid lines, Wilson equation. (a, b, c) Curves refer to dimethyl carbonate + methanol, + ethanol, and + propan-1-ol mixtures.

The experimental values x_i, y_i, and P are listed in Table 3, reporting also the "experimental" activity coefficients γ_i, which were evaluated by the equation relating the P, T, x_i, and y_i values

$$\gamma_i = Py_i/P_ix_i \qquad i = 1, 2 \tag{2}$$

with P_i obtained from $P°_i$ and a fugacity factor (Van Ness, 1995).

Graphical representations of the VLE measurements are shown in Figure 1.

The activity coefficients γ_i were fitted to Wilson equation

$$\ln \gamma_k = -\ln(x_k - \Lambda_{kj}x_j) + x_j[\Lambda_{kj}/(x_k + \Lambda_{kj}x_j) - a_{jk}/(x_j + \Lambda_{jk}x_k)] \tag{3}$$

$$\Lambda_{ij} = (V_j/V_i)\exp(-a_{ij}/RT); \quad k = 1, 2; \ k \neq j$$

through the least-squares procedure used in a previous paper (Francesconi et al., 1993).

The parameters a_{ij}, together with standard deviations σ, are listed in Table 6.

The Wilson equation (eq 3) was also employed to calculate the activity coefficients at infinite dilution, $\gamma_i^\infty = \lim_{x_i \to 0} \gamma_i$, reported in Table 3.

We have used other models, like the ones of NRTL (Renon and Prausnitz, 1969) and Redlich−Kister (Carra'

Table 3. Pressures, Mole Fractions, x_1 and y_1, in Liquid and Vapor Phases, and Experimental Activity Coefficients, γ_i, for Dimethyl Carbonate + 1-Alkanol Mixtures at 313.15 K

P/kPa	x_1	y_1	γ_1	γ_2
Dimethyl Carbonate (1) + Methanol (2)				
36.40	0.012	0.024	4.59	1.02
36.80	0.016	0.030	4.50	1.03
37.00	0.024	0.042	4.28	1.03
37.40	0.036	0.061	4.14	1.03
37.80	0.050	0.079	3.92	1.04
38.05	0.068	0.099	3.65	1.04
38.20	0.088	0.119	3.38	1.04
38.35	0.106	0.134	3.19	1.05
38.40	0.124	0.148	3.00	1.06
38.55	0.146	0.163	2.82	1.07
38.65	0.173	0.177	2.59	1.09
38.60	0.206	0.194	2.38	1.11
38.50	0.246	0.212	2.17	1.14
38.30	0.297	0.231	1.95	1.19
38.00	0.354	0.248	1.75	1.25
37.40	0.424	0.265	1.54	1.35
36.75	0.479	0.289	1.45	1.42
35.60	0.576	0.318	1.29	1.62
35.80	0.631	0.343	1.28	1.81
32.70	0.720	0.385	1.15	2.03
29.90	0.797	0.432	1.07	2.37
26.70	0.868	0.508	1.03	2.82
22.70	0.927	0.634	1.03	3.22
20.20	0.958	0.741	1.03	3.57
19.70	0.961	0.759	1.03	3.44
19.20	0.967	0.789	1.03	3.46
18.00	0.978	0.838	1.02	3.75
$\gamma_1^\infty = 4.66$, $\gamma_2^\infty = 3.80$, eq 3. $I = -0.003$, $\delta I = 0.20$, eq 5.				
Dimethyl Carbonate (1) + Ethanol (2)				
19.20	0.015	0.067	5.81	1.02
19.60	0.021	0.090	5.48	1.02
20.05	0.032	0.126	5.24	1.02
20.40	0.044	0.153	4.66	1.01
20.65	0.053	0.173	4.42	1.01
21.10	0.068	0.204	4.17	1.01
21.90	0.101	0.251	3.61	1.02
22.20	0.125	0.271	3.19	1.04
22.40	0.137	0.284	3.07	1.04
22.70	0.156	0.300	2.89	1.06
23.20	0.201	0.332	2.54	1.09
23.50	0.237	0.350	2.29	1.12
23.70	0.281	0.376	2.10	1.15
23.90	0.338	0.399	1.86	1.22
24.00	0.421	0.429	1.61	1.33
23.95	0.472	0.446	1.50	1.41
23.90	0.503	0.456	1.43	1.47
23.65	0.561	0.475	1.32	1.58
23.60	0.573	0.483	1.31	1.60
23.40	0.637	0.508	1.23	1.78
23.15	0.661	0.522	1.21	1.83
22.10	0.785	0.597	1.11	2.33
21.30	0.837	0.646	1.09	2.59
20.60	0.874	0.676	1.05	2.97
19.30	0.911	0.741	1.04	3.17
18.40	0.938	0.801	1.04	3.34
17.60	0.957	0.848	1.03	3.51
16.9	0.972	0.890	1.03	3.71
$\gamma_1^\infty = 5.90$, $\gamma_2^\infty = 3.90$, eq 3. $I = 0.041$, $\delta I = 0.16$, eq 5.				
Dimethyl Carbonate (1) + Propan-1-ol (2)				
8.70	0.025	0.240	5.52	0.98
10.40	0.064	0.358	3.85	1.03
12.20	0.125	0.474	3.07	1.06
13.30	0.181	0.532	2.59	1.09
14.20	0.240	0.578	2.27	1.13
14.80	0.313	0.614	1.93	1.19
15.30	0.392	0.644	1.67	1.29
15.60	0.437	0.657	1.56	1.37
15.95	0.502	0.676	1.42	1.49
16.10	0.552	0.694	1.34	1.58
16.30	0.601	0.707	1.27	1.72
16.45	0.661	0.726	1.20	1.91
16.50	0.724	0.760	1.15	2.06
16.55	0.762	0.781	1.12	2.19
16.60	0.807	0.803	1.10	2.43
16.55	0.845	0.827	1.07	2.66
16.45	0.876	0.844	1.05	2.97
16.40	0.901	0.865	1.04	3.21
16.25	0.930	0.900	1.04	3.32
16.15	0.950	0.925	1.04	3.46
$\gamma_1^\infty = 5.57$, $\gamma_2^\infty = 3.81$, eq 3. $I = 0.021$, $\delta I = 0.11$, eq 5.				

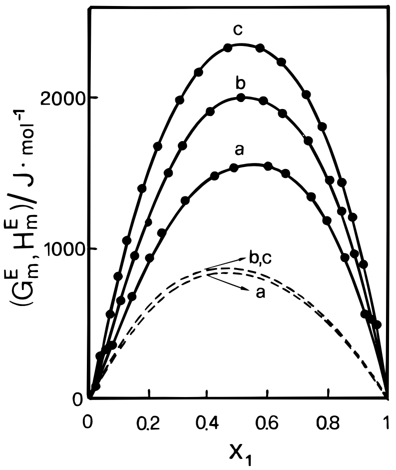

Figure 2. Excess molar enthalpies, H_m^E, and excess molar Gibbs energies, G_m^E (dashed curves), eq 3, for dimethyl carbonate (1) + 1-alkanol (2) mixtures at 313.15 K. (●) Experimental points; solid lines, Redlich−Kister equation. (a, b, c) Curves refer to dimetyl carbonate + methanol, + ethanol, and + propan-1-ol.

Table 4. Mole Fractions, x_1, and Experimental Excess Molar Enthalpies, H_m^E, of Dimethyl Carbonate + 1-Alkanols at 313.15 K

x_1	H_m^E/J·mol^{-1}	x_1	H_m^E/J·mol^{-1}
Dimethyl Carbonate (1) + Methanol (2)			
0.0196	82	0.4901	1537
0.0385	168	0.5905	1555
0.0741	323	0.6578	1504
0.1381	660	0.7425	1342
0.1937	905	0.7936	1189
0.2426	1085	0.8522	947
0.3246	1308	0.9202	562
0.4189	1474		
Dimethyl Carbonate (1) + Ethanol (2)			
0.0280	159	0.5090	1975
0.0544	333	0.5802	1966
0.1033	650	0.6746	1861
0.1473	954	0.7343	1707
0.1872	1167	0.8057	1444
0.2568	1499	0.8468	1232
0.3154	1700	0.8924	949
0.4086	1890	0.9431	542
Dimethyl Carbonate (1) + Propan-1-ol (2)			
0.0355	287	0.5756	2315
0.0686	561	0.6387	2235
0.0995	811	0.7261	2021
0.1284	1033	0.7795	1819
0.1810	1399	0.8414	1435
0.2275	1682	0.8761	1211
0.3065	1985	0.9138	883
0.3707	2163	0.9550	485
0.4691	2314		

et al., 1969) with more parameters, but the best fits were obtained from Wilson model.

The excess molar Gibbs energies, G_m^E, graphically represented (dashed curves) in Figure 2, were calculated from

$$G_m^E = RT(x_1 \ln \gamma_1 + x_2 \ln \gamma_2) \qquad (4)$$

with γ_i given by eq 3.

The thermodynamic consistency of the VLE data was tested following the procedure described in previous papers (Francesconi and Comelli, 1993; Comelli and Francesconi, 1996).

Table 3 reports values of area I and its uncertainties δI, with

$$I = \int_0^1 [\ln(\gamma_1/\gamma_2) + V_m^E/RT \cdot dP/dx]\, dx \qquad (5)$$

Table 5. Mole Fractions, x_1, Experimental Densities, ρ, and Excess Molar Volumes, V^{E}_{m}, of Dimethyl Carbonate + 1-Alkanols at 313.15 K

x_1	ρ/g·cm^{-3}	V^{E}_{m}/ cm^{3}·mol^{-1}	x_1	ρ/g·cm^{-3}	V^{E}_{m}/ cm^{3}·mol^{-1}
		Dimethyl Carbonate (1) + Methanol (2)			
0.0109	0.778 43	−0.003	0.5321	0.963 48	−0.041
0.0526	0.800 61	−0.013	0.6126	0.980 70	−0.034
0.0942	0.820 90	−0.020	0.7259	1.001 98	−0.016
0.1166	0.831 17	−0.025	0.7656	1.008 75	−0.009
0.1700	0.853 88	−0.031	0.8106	1.016 06	−0.003
0.2152	0.871 41	−0.036	0.8305	1.019 17	0.002
0.2724	0.891 70	−0.042	0.8757	1.026 01	0.006
0.2952	0.899 26	−0.044	0.8983	1.029 31	0.009
0.3277	0.909 54	−0.045	0.9374	1.034 87	0.009
0.3569	0.918 32	−0.046	0.9575	1.037 65	0.007
0.4075	0.932 61	−0.047	0.9734	1.039 81	0.006
0.4523	0.944 38	−0.046	0.9745	1.039 97	0.005
		Dimethyl Carbonate (1) + Ethanol (2)			
0.0242	0.781 46	0.025	0.4851	0.925 84	0.221
0.0589	0.794 01	0.057	0.5527	0.943 32	0.222
0.1309	0.819 32	0.109	0.6076	0.956 94	0.221
0.1546	0.827 35	0.124	0.6507	0.967 36	0.213
0.2348	0.853 51	0.161	0.7131	0.981 92	0.200
0.2720	0.865 10	0.177	0.7277	0.985 26	0.195
0.3355	0.884 18	0.195	0.8088	1.003 32	0.163
0.3764	0.896 01	0.205	0.8950	1.021 94	0.107
0.4526	0.917 16	0.217	0.9641	1.036 09	0.042
		Dimethyl Carbonate (1) + Propan-1-ol (2)			
0.0363	0.797 02	0.072	0.5492	0.930 82	0.430
0.0567	0.802 46	0.109	0.5909	0.941 37	0.421
0.1407	0.824 86	0.232	0.6846	0.964 88	0.380
0.2324	0.849 12	0.327	0.7127	0.971 90	0.364
0.2768	0.860 76	0.360	0.7522	0.981 71	0.334
0.3283	0.874 20	0.390	0.8361	1.002 58	0.253
0.3854	0.889 01	0.414	0.8827	1.014 16	0.195
0.4337	0.901 43	0.427	0.9364	1.027 51	0.115
0.5143	0.922 01	0.432	0.9641	1.034 43	0.068

For the three mixtures, $\delta I > |I|$, and then the consistency is verified.

The term containing V^{E}_{m} in eq 4, evaluated from density measurements, Table 5, plays a negligible role in the calculation of I.

The H^{E}_{m} and V^{E}_{m} values, listed in Tables 4 and 5, graphically represented in Figures 2 and 3 respectively, have been fitted to the Redlich−Kister equation

$$Q^{E}_{m} = x_1 x_2 \sum_{k \geq 0} a_k (x_1 - x_2)^k \qquad (6)$$

where $Q^{E}_{m} = H^{E}_{m}$/J·mol^{-1} or V^{E}_{m}/cm^{3}·mol^{-1}, by a least-squares method with all points weighted equally.

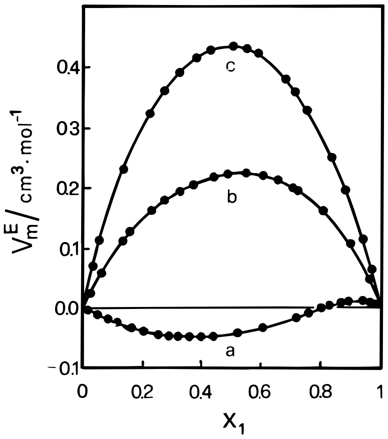

Figure 3. Excess molar volumes, V^{E}_{m}, for dimethyl carbonate (1) + 1-alkanol (2) mixtures at 313.15 K. (●) Experimental points; solid lines, Redlich−Kister equation. (a, b, c) Curves refer to dimethyl carbonate + methanol, + ethanol, and + propan-1-ol mixtures, respectively.

The adjustable parameters a_k and the standard deviation $\sigma(Q^{E}_{m})$

$$\sigma(Q^{E}_{m}) = |\Phi/(N - n)|^{0.5} \qquad (7)$$

with N = number of experimental points and n = number of adjustable parameters, are showed in Table 6.

Φ is the objective function defined as

$$\Phi = \sum_{k \geq 0}^{N} \eta_k^2 \qquad (8)$$

where $\eta = Q^{E}_{m,calcd} - Q^{E}_{m}$, $Q^{E}_{m,calcd}$ being determined from the right-hand side of eq 6.

All mixtures are strongly endothermic; the enthalpy values increase with the increase of the chain length of alkanol showing maxima varying from 1560 (methanol) to 2340 (propan-1-ol) J·mol^{-1}. The V^{E}_{m}'s are positive over the entire composition range for dimethyl carbonate + ethanol and + propan-1-ol mixtures, showing and inversion of sign for dimethyl carbonate + methanol.

The G^{E}_{m} curves for the three alkanols are almost coincident, whereas the H^{E}_{m} curves are nearly equispaced, by about 200 J·mol^{-1}. Hence, also the excess entropy term $TS^{E}_{m} = H^{E}_{m} - G^{E}_{m}$ increases regularly in passing from methanol to propan-1-ol.

Most of the conclusions drawn on the mixtures 1,2-epoxybutane + alkanols (Francesconi and Comelli, 1996)

Table 6. Least-Squares Parameters, a_k, and Standard Deviations, σ, Eqs 3 and 6, for Dimethyl Carbonate + 1-Alkanol Mixtures at 313.15 K

function	a_0	a_1	a_2	a_3	a_4	σ
			Dimethyl Carbonate + Methanol			
G^{E}_{m} [a]/J·mol^{-1}	1749.1	2750.2				0.02
H^{E}_{m} [b]/J·mol^{-1}	6170.8	957.2	1915.8	856.7	−2425.6	5.9
V^{E}_{m} [b]/cm^{3}·mol^{-1}	−0.1772	0.1113	0.1582	0.1547		0.0005
			Dimethyl Carbonate + Ethanol			
G^{E}_{m} [a]/J·mol^{-1}	2440.7	2606.6				0.04
H^{E}_{m} [b]/J·mol^{-1}	7886.6	595.0	2555.9	1783.7	−2581.3	7.4
V^{E}_{m} [b]/cm^{3}·mol^{-1}	0.8845	0.1061	0.2776			0.0009
			Dimethyl Carbonate + Propan-1-ol			
G^{E}_{m} [a]/J·mol^{-1}	2944.2	1944.7				0.05
H^{E}_{m} [b]/J·mol^{-1}	9285.9	606.9	3190.0	1071.8	−2498.3	10.5
V^{E}_{m} [b]/cm^{3}·mol^{-1}	1.7290	−0.0109	0.3119	−0.0539		0.0008

[a] Parameters used to calculate G^{E}_{m}, eq 4, and obtained from a best fit of the γ_i values of eq 3, through a Wilson equation, with a_0, a_1 = a_{12}, a_{21}. [b] Parameters obtained from a best fit of calorimetric and volumetric data through the Redlich−Kister expression, eq 6.

may be shared by the mixtures of the present paper, which, however, have this peculiar feature: the values of H_m^E and V_m^E are greater, the azeotropes are more marked, and the activity coefficients range up to 3 (for the mixtures containing 1,2-epoxybutane, maximum values of activity coefficients are around 1.3).

All these results lead to the conclusion that deviations from ideality are larger for the mixtures of this paper and are certainly due to both self-association of alkanols and formation of associates by charge transfer between the OH group of the alkanol and the C═O group of the ester.

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