Densities, Relative Permittivities, and Refractive Indices of the Binary Systems Propylene Carbonate + *o*-Xylene and Propylene Carbonate + *m*-Xylene at (15, 20, 25, 30, and 35) °C

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Densities, relative permittivities, and refractive indices were measured at (15, 20, 25, 30, and 35) °C, over the whole composition range of the binary mixtures propylene carbonate + *o*-xylene and propylene carbonate + *m*-xylene. Excess volumes were found to be negative, and the values were fitted to a Redlich−Kister type equation. Apparent dipole moments were calculated, as well as deviations in relative permittivity, molar refraction, and molar polarization.

Introduction

Propylene carbonate is an aprotic solvent with high relative permittivity, is a liquid in a wide temperature range, is very popular for electrochemical investigations, and is widely used as a nonaqeous electrolyte in high-energy lithium batteries (Gabano, 1983). The physicochemical properties and intermolecular interactions in mixtures of propylene carbonate with aromatic hydrocarbons are widely studied in our laboratory (Moumouzias and Ritzoulis, 1992; Moumouzias and Ritzoulis, 1997).

The binary system propylene carbonate + *p*-xylene was recently investigated (Konti et al., 1997), whereas our study in binary mixtures of propylene carbonate with xylenes is integrated here by presenting the results of density, relative permittivity, and refractive index measurements of propylene carbonate + *o*-xylene and propylene carbonate + *m*-xylene mixtures, from (15 to 35) °C. Excess volumes were calculated, as well as deviations in relative permittivity, molar refraction, and molar polarization. Apparent dipole moments were also calculated.

Experimental Section

o-Xylene and *m*-xylene (Fluka, puriss p.a. >99%) were distilled, and the middle fractions were collected. Propylene carbonate (Merck, >99%) was dried with molecular sieves (5 Å) and fractionally distilled under reduced pressure. The solutions were prepared by mass by means of an analytical balance of four decimal places. The error in the mole fraction was estimated to be lower than ±0.000 03.

A dipolmeter of Wissenschaftlich Technische Werkstätten GmbH, model DM-01, was used for relative permittivity measurements, and the uncertainty in ϵ was <0.3%. Refractive indices were measured by means of an Abbe refractometer (aus JENA, model G) for sodium light. The accuracy was better than ±0.0002 units. The temperature in the dipolmeter and the refractometer was regulated with a YSI (model 72) thermostat with a stability of ±0.005 °C.

Density measurements were made with an Anton−Paar DMA 602/60 vibrating tube densimeter, thermostated with a Haake F3-K Digital thermostat with a stability of ±0.02 °C. The accuracy in density was better than $\pm 5 \times 10^{-5}$ g cm^{-3}. More details about the experimental conditions are given elsewhere (Konti et al., 1997).

Results and Discussion

The experimental results of density, relative permittivity, and refractive index are given in Tables 1 and 2 for propylene carbonate + *o*-xylene and propylene carbonate + *m*-xylene systems, correspondingly. The experimental and the literature values of the pure components are given in Table 3. As can be seen, the experimental values generally are in agreement with the literature ones. The refractive index experimental value of propylene carbonate at 25 °C is 1.4194. This value is lower than the corresponding one found by Pal et al. (1998) 1.4210, but the experimental value at 20 °C, 1.4214, is in absolute agreement with the value reported by Jorat et al. (1991).

Density measurements were used for the calculation of excess volume V^E from the equation

$$V^{E} = M_1x_1\left(\frac{1}{d} - \frac{1}{d_1}\right) + M_2x_2\left(\frac{1}{d} - \frac{1}{d_2}\right) \qquad (1)$$

In the above equation d is the density of the mixture and d_i, M_i, and x_i are, correspondingly, the density, the molar mass, and the mole fraction of the pure components. Variations of excess volume with the mole fraction of propylene carbonate are given in Figures 1 and 2. In both systems, all the V^E values are found to be negative, with minimums at $x_1 \approx 0.3-0.4$, and become more negative as the temperature increases. This behavior has also been observed in propylene carbonate + *p*-xylene systems (Konti et al., 1997) as well as in other binary mixtures of propylene carbonate (Gopal and Agarwal, 1976; Moumouzias and Ritzoulis, 1992; Moumouzias et al., 1991). The negative values of V^E can be explained by the different geometries of propylene carbonate and xylenes molecules or by dipole−induced dipole interactions between them (Fort and Moore, 1965; Palepu et al., 1987; Prigogine, 1957). According to Joshi et al. (1990), the large difference between the relative permittivities of the unlike components could also be responsible for the observed negative values of V^E.

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Table 1. Experimental Values of Density d, Relative Permittivity ϵ, and Refractive Index n_D for Different Values of Mole Fraction x_1 for Propylene Carbonate (1) + *o*-Xylene (2) Systems

	d/g cm^{-3}				
x_1	15 °C	20 °C	25 °C	30 °C	35 °C
0.0000	0.8853	0.8808	0.8762	0.8715	0.8671
0.1033	0.9106	0.9061	0.9014	0.8967	0.8922
0.2084	0.9374	0.9328	0.9282	0.9234	0.9189
0.3078	0.9641	0.9595	0.9547	0.9499	0.9453
0.4114	0.9936	0.9889	0.9841	0.9792	0.9746
0.5111	1.0239	1.0192	1.0142	1.0093	1.0046
0.6089	1.0557	1.0508	1.0458	1.0408	1.0360
0.7088	1.0906	1.0856	1.0804	1.0754	1.0706
0.8066	1.1274	1.1223	1.1170	1.1119	1.1069
0.9053	1.1674	1.1621	1.1569	1.1516	1.1465
1.0000	1.2088	1.2035	1.1981	1.1927	1.1875

	ϵ				
x_1	15 °C	20 °C	25 °C	30 °C	35 °C
0.0000	2.60	2.58	2.56	2.55	2.53
0.1033	5.37	5.39	5.32	5.22	5.14
0.2084	9.11	9.07	8.93	8.78	8.65
0.3078	13.50	13.34	13.12	12.92	12.74
0.4114	18.96	18.64	18.34	18.03	17.77
0.5111	25.07	24.59	24.18	23.74	23.37
0.6089	31.87	31.25	30.73	30.11	29.61
0.7088	39.64	38.90	38.26	37.45	36.78
0.8066	48.07	47.21	46.45	45.49	44.65
0.9053	57.38	56.40	55.55	54.50	53.53
1.0000	67.09	65.96	65.05	64.08	63.02

	n_D				
x_1	15 °C	20 °C	25 °C	30 °C	35 °C
0.0000	1.5079	1.5053	1.5028	1.5003	1.4978
0.1033	1.5017	1.4992	1.4967	1.4943	1.4918
0.2084	1.4948	1.4924	1.4900	1.4876	1.4852
0.3078	1.4878	1.4854	1.4830	1.4807	1.4784
0.4114	1.4798	1.4776	1.4752	1.4730	1.4707
0.5111	1.4717	1.4696	1.4673	1.4651	1.4629
0.6089	1.4634	1.4612	1.4589	1.4568	1.4547
0.7088	1.4541	1.4521	1.4499	1.4478	1.4458
0.8066	1.4445	1.4426	1.4404	1.4384	1.4363
0.9053	1.4340	1.4321	1.4301	1.4281	1.4261
1.0000	1.4232	1.4214	1.4194	1.4174	1.4155

Table 2. Experimental Values of Density d, Relative Permittivity ϵ, and Refractive Index n_D for Different Values of Mole Fraction x_1 for Propylene Carbonate (1) + *m*-Xylene (2) Systems

	d/g cm^{-3}				
x_1	15 °C	20 °C	25 °C	30 °C	35 °C
0.0000	0.8684	0.8642	0.8602	0.8553	0.8521
0.1040	0.8948	0.8905	0.8864	0.8815	0.8782
0.2068	0.9221	0.9177	0.9136	0.9086	0.9051
0.3063	0.9500	0.9455	0.9413	0.9362	0.9327
0.4070	0.9799	0.9754	0.9710	0.9660	0.9622
0.5101	1.0128	1.0082	1.0036	0.9986	0.9946
0.6125	1.0479	1.0432	1.0385	1.0334	1.0292
0.7060	1.0824	1.0776	1.0727	1.0676	1.0632
0.8091	1.1234	1.1184	1.1134	1.1083	1.1035
0.9014	1.1631	1.1580	1.1527	1.1475	1.1425
1.0000	1.2088	1.2035	1.1981	1.1927	1.1875

	ϵ				
x_1	15 °C	20 °C	25 °C	30 °C	35 °C
0.0000	2.43	2.41	2.39	2.37	2.35
0.1040	5.36	5.28	5.20	5.10	5.03
0.2068	8.87	8.73	8.60	8.44	8.31
0.3063	13.10	12.88	12.69	12.47	12.29
0.4070	18.30	17.98	17.70	17.41	17.16
0.5101	24.57	24.12	23.73	23.33	22.98
0.6125	31.71	31.09	30.58	30.02	29.53
0.7060	38.98	38.18	37.55	36.82	36.18
0.8091	47.84	46.84	46.08	45.18	44.36
0.9014	56.60	55.48	54.63	53.63	52.66
1.0000	67.09	65.96	65.05	64.08	63.02

	n_D				
x_1	15 °C	20 °C	25 °C	30 °C	35 °C
0.0000	1.4997	1.4973	1.4945	1.492	1.4896
0.1040	1.4942	1.4918	1.4892	1.4867	1.4844
0.2068	1.4883	1.4860	1.4834	1.4810	1.4787
0.3063	1.4820	1.4797	1.4772	1.4749	1.4726
0.4070	1.4752	1.4729	1.4705	1.4683	1.4660
0.5101	1.4677	1.4655	1.4631	1.4610	1.4588
0.6125	1.4597	1.4575	1.4552	1.4532	1.4510
0.7060	1.4518	1.4497	1.4475	1.4455	1.4433
0.8091	1.4425	1.4404	1.4383	1.4363	1.4342
0.9014	1.4335	1.4315	1.4295	1.4274	1.4255
1.0000	1.4232	1.4214	1.4194	1.4174	1.4155

To evaluate deviations in relative permittivity, we used the expression

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

where ϵ is the relative permittivity of the mixture, ϵ_i is the relative permittivity of the pure components, $\Delta\epsilon$ is the deviation in the relative permittivity, and φ_i is the volume fraction (Böttcher, 1973; Buep and Barón, 1988; Nath and Narain, 1982; Jannelli et al., 1983). The temperature effect in both systems is not important, and the variation of $\Delta\epsilon$ with the volume fraction of propylene carbonate at 25 °C is presented in Figure 3. As can be seen, the behavior of $\Delta\epsilon$ is similar in the two systems, as well as in the propylene carbonate + *p*-xylene system (Konti et al., 1997); $\Delta\epsilon$ values are negative, and the largest deviations are about −5, corresponding at $\varphi_1 \approx 0.4$.

Table 3. Experimental and Literature Values of Physical Properties of the Pure Components at 25 °C

	d/g cm^{-3}		ϵ		n_D	
	exp	lit.	exp	lit.	exp	lit.
propylene carbonate	1.1981	1.1970[a] 1.1980[b] 1.1989[c] 1.1995[d]	65.05	65.1[a] 65.04[c] 64.92[e]	1.4194 1.4214[n]	1.4210[f] 1.4214[g,n]
o-xylene	0.8762	0.87582[h] 0.87558[i] 0.87563[j]	2.56		1.5028	1.50252[h] 1.50268[i] 1.50177[k]
m-xylene	0.8602	0.85976[h] 0.86006[i] 0.85986[j] 0.8602[l] 0.8596[m]	2.39		1.4945	1.49416[h] 1.49466[i] 1.49443[k] 1.4945[l]

[a] Kronick and Fuoss, 1955. [b] Wu and Friedman, 1966. [c] Hanna and Al-Sudani, 1987. [d] Salomon, 1969. [e] Payne and Theodorou, 1972. [f] Pal et al., 1998. [g] Jorat et al., 1991. [h] Tanaka et al., 1975. [i] Serrano et al., 1990. [j] TRC, 1998. [k] Tojo and Diaz, 1995. [l] Aralaguppi et al., 1992. [m] Chang and Lee, 1995. [n] At 20 °C.

Molar refraction was calculated from refractive index data, according to the Lorentz−Lorenz relation (Minkin et al., 1970)

$$[R] = \frac{{n_D}^2 - 1}{{n_D}^2 + 2}\frac{M}{d} \tag{3}$$

[R], ${n_D}^2$, and M are, correspondingly, the molar refraction, the refractive index, and the molar mass of the mixture. Deviations in molar refraction were calculated from

$$\Delta[R] = [R] - (x_1[R]_1 + x_2[R]_2) \tag{4}$$

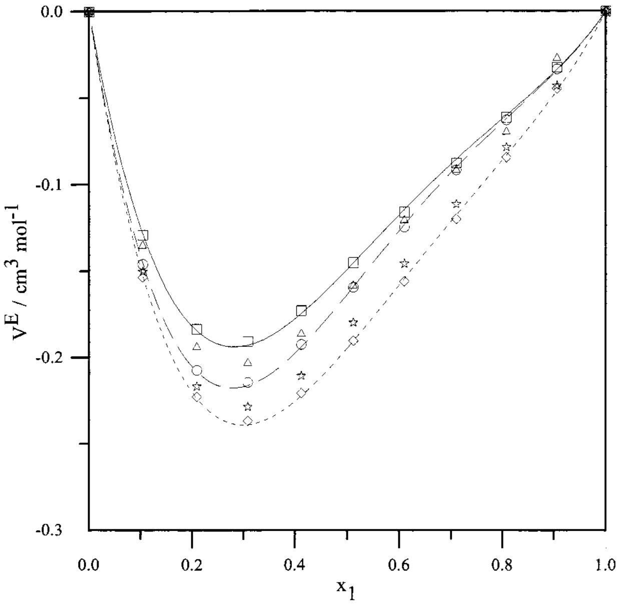

Figure 1. Dependence of excess volume V^E on the mole fraction x_1 for propylene carbonate (1) + *o*-xylene (2) systems: □, 15 °C; Δ, 20 °C; ○, 25 °C; ☆, 30 °C; ◇, 35 °C. Curves are least-squares representations by eq 7a (—, 15 °C; — — —, 25 °C; - - -, 35 °C).

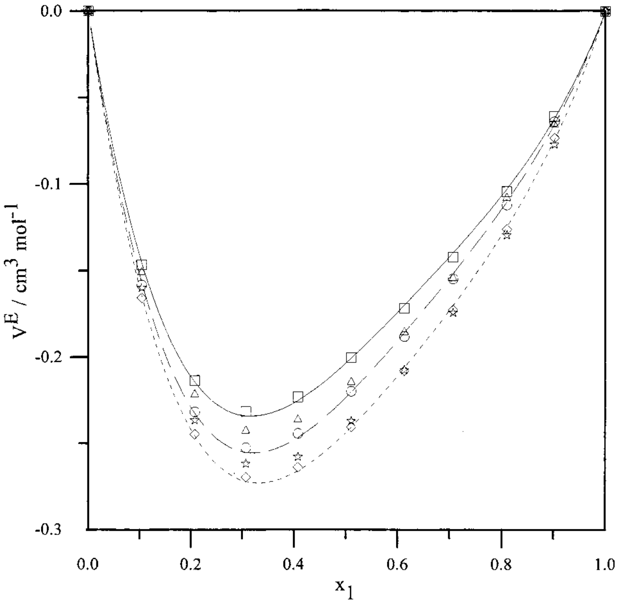

Figure 2. Dependence of excess volume V^E on the mole fraction x_1 for propylene carbonate (1) + *m*-xylene (2) systems: □, 15 °C; Δ, 20 °C; ○, 25 °C; ☆, 30 °C; ◇, 35 °C. Curves are least-squares representations by eq 7a (—, 15 °C; — — —, 25 °C; - - -, 35 °C).

where $[R]_1$ and $[R]_2$ refer to the pure components. The temperature effect was not important, and in both systems, $\Delta[R]$ is negative with a minimum at $x_1 \approx 0.3-0.4$, like the V^E-x_1 curves. $\Delta[R]$ absolute values are a little higher for the propylene carbonate + *m*-xylene system. Variations of $\Delta[R]$ with mole fraction of propylene carbonate for the two systems at 25 °C are presented in Figure 4.

The Kirkwood−Frohlich expression (Nath and Tripathi, 1984; Ruostesuo and Liias-Lepisto, 1991)

$$P_m = \frac{(\epsilon - n_D^{\ 2})(2\epsilon + n_D^{\ 2})}{9\epsilon}V \tag{5}$$

was used for the calculation of the molar polarization P_m, whereas deviations of the molar polarization ΔP_m were calculated from

$$\Delta P_m = P_m - (x_1 P_{m,1} + x_2 P_{m,2}) \tag{6}$$

where $P_{m,i}$ are the molar polarizations of the pure components. ΔP_m was not found to be sensitive to temperature variations, and ΔP_m values, at 25 °C, are plotted against x_1 in Figure 5. As one can see, the two systems show exactly the same behavior: negative values in the entire range with a minimum at $x_1 \approx 0.4$.

The values of the excess volumes, the relative permittivity, the molar refraction, and the molar polarization

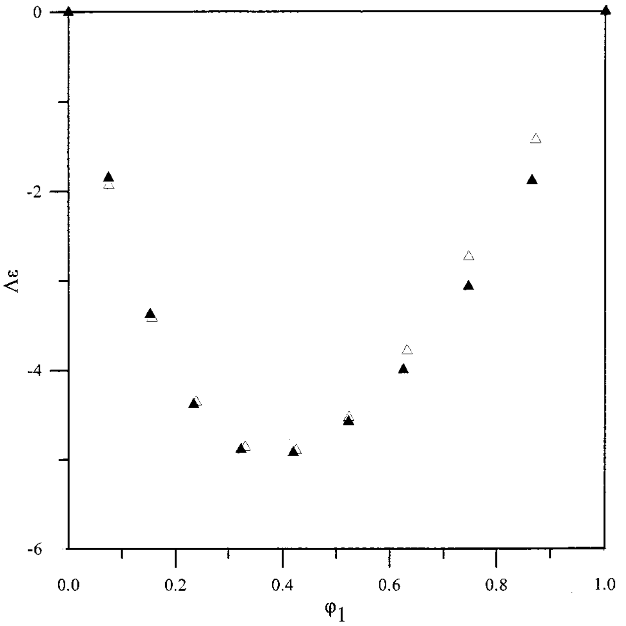

Figure 3. Dependence of the deviation in relative permittivity $\Delta\epsilon$ on the volume fraction φ_1 for propylene carbonate (1) + *o*-xylene (2) (open symbols) and propylene carbonate (1) + *m*-xylene (2) (full symbols) systems, at 25 °C.

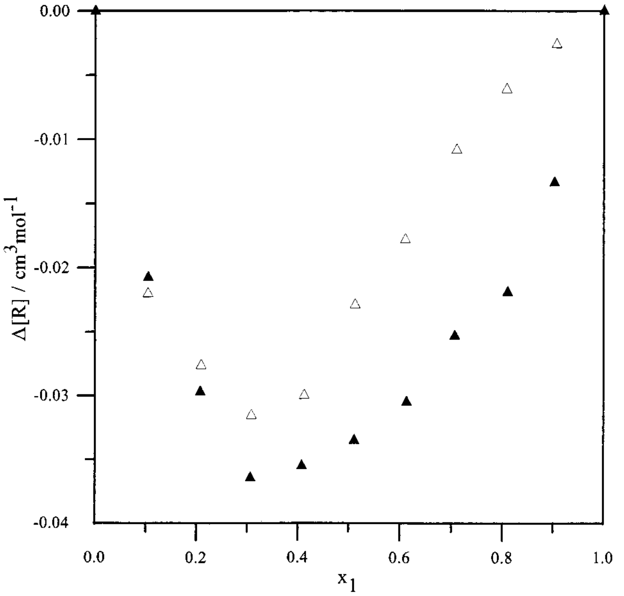

Figure 4. Dependence of the deviation in molar refraction $\Delta[R]$ on the mole fraction x_1 for propylene carbonate (1) + *o*-xylene (2) (open symbols) and propylene carbonate (1) + *m*-xylene (2) (full symbols) systems, at 25 °C.

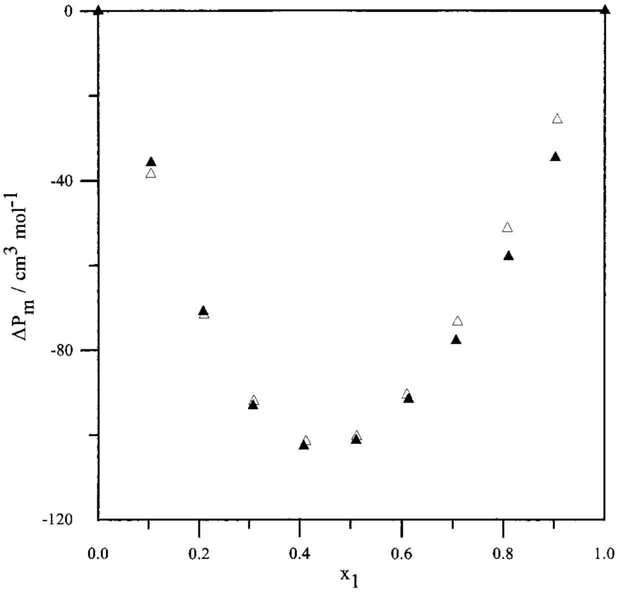

Figure 5. Dependence of the deviation in molar polarization ΔP_m on the mole fraction x_1 for propylene carbonate (1) + *o*-xylene (2) (open symbols) and propylene carbonate (1) + *m*-xylene (2) (full symbols) systems, at 25 °C.

Table 4. Coefficients a_j, b_j, c_j, and d_j and Standard Deviations σ (cm^3 mol^{-1}) of Eq 7 for Propylene Carbonate + *o*-Xylene Systems

	15 °C	20 °C	25 °C	30 °C	35 °C
			Eq 7a		
a_0	−0.596	−0.647	−0.654	−0.735	−0.778
a_1	0.627	0.596	0.738	0.698	0.702
a_2	−0.463	−0.413	−0.522	−0.520	−0.495
a_3		0.176			
σ	0.010	0.029	0.008	0.007	0.011
			Eq 7b		
b_0	−18.43	−18.47	−18.53	−18.79	−18.46
b_1	10.51	9.71	9.28	8.00	7.40
b_2	−3.23	−1.77	−1.87	−3.09	−3.73
σ	0.10	0.01	0.01	0.14	0.19
			Eq 7c		
c_0	−0.079	−0.076	−0.094	−0.094	−0.093
c_1	0.113	0.103	0.123	0.125	0.138
c_2	−0.064	−0.028	−0.055	−0.048	−0.061
σ	0.011	0.005	0.009	0.004	0.012
			Eq 7d		
d_0	−402.7	−401.9	−403.8	−407.2	−399.7
d_1	130.0	111.5	102.3	73.16	61.92
d_2	26.78	49.90	46.85	11.06	−5.73
d_3	−44.73	−50.22	−49.88	−41.27	−38.91
d_4	34.88	41.06	37.31	33.58	30.35
σ	0.67	0.64	0.76	0.70	0.72

deviations were fitted to smoothing Redlich−Kister type equations (Redlich and Kister, 1948)

$$V^E = x_1x_2\sum_{j=0} a_j(x_1 - x_2)^j \quad (7a)$$

$$\Delta\epsilon = \varphi_1\varphi_2\sum_{j=0} b_j(\varphi_1 - \varphi_2)^j \quad (7b)$$

$$\Delta[R] = x_1x_2\sum_{j=0} c_j(x_1 - x_2)^j \quad (7c)$$

$$\Delta P_m = x_1x_2\sum_{j=0} d_j(x_1 - x_2)^j \quad (7d)$$

The coefficients a_j, b_j, c_j, and d_j were evaluated from a least-squares method and are presented in Table 4 for propylene carbonate + *o*-xylene systems and in Table 5 for propylene carbonate + *m*-xylene.

Table 5. Coefficients a_j, b_j, c_j, and d_j and Standard Deviations σ (cm^3 mol^{-1}) of Eq 7 for Propylene Carbonate + *m*-xylene Systems

	15 °C	20 °C	25 °C	30 °C	35 °C
			Eq 7a		
a_0	−0.814	−0.869	−0.893	−0.959	−0.974
a_1	0.547	0.550	0.600	0.525	0.588
a_2	−0.501	−0.485	−0.503	−0.527	−0.519
σ	0.012	0.014	0.015	0.009	0.012
			Eq 7b		
b_0	−18.71	−18.70	−18.78	−18.83	−18.60
b_1	10.82	9.66	8.98	7.83	7.1
b_2	−3.36	−4.15	−4.31	−5.45	−6.01
b_3	−4.64	−4.44	−4.09	−3.80	−3.43
σ	0.23	0.24	0.24	0.25	0.24
			Eq 7c		
c_0	−0.112	−0.141	−0.135	−0.109	−0.130
c_1	0.071	0.047	0.045	0.041	0.043
c_2	−0.118	−0.157	−0.081	−0.181	−0.104
σ	0.007	0.015	0.006	0.015	0.010
			Eq 7d		
d_0	−408.5	−405.8	−407.1	−406.4	−398.9
d_1	137.3	120.2	107.9	93.16	84.29
d_2	27.26	−0.46	−11.68	−46.85	−65.70
d_3	−171.9	−180.2	−176.5	−186.8	−187.4
d_4	66.29	68.49	70.82	71.99	72.15
σ	1.19	1.23	1.08	1.07	1.03

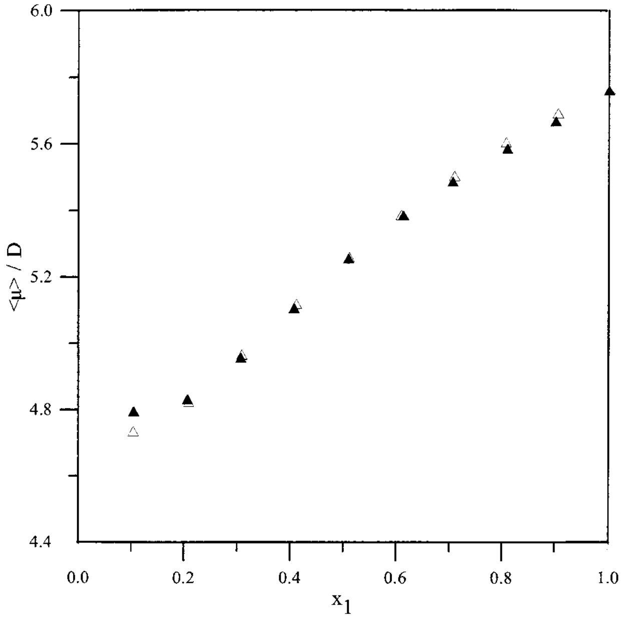

Figure 6. Dependence of $\langle\mu\rangle$ on the mole fraction x_1 for propylene carbonate (1) + *o*-xylene (2) (open symbols) and propylene carbonate (1) + *m*-xylene (2) (full symbols) systems, at 25 °C.

The apparent dipole moment $\langle\mu\rangle$ of a polar solute in a nonpolar solvent can be calculated from the Kirkwood−Fröhlich equation (Böttcher, 1973)

$$\langle\mu\rangle^2 = \frac{9kT(2\epsilon + \epsilon_{\infty,1})^2}{4\pi N_A x_1(\epsilon_{\infty,1} + 2)^2(2\epsilon + 1)} \times \left[\frac{V(\epsilon - 1)}{\epsilon} - \frac{3(1 - x_1)V_2(\epsilon_2 - 1)}{(2\epsilon + \epsilon_2)} - \frac{3x_1V_1(\epsilon_{\infty,1} - 1)}{(2\epsilon + \epsilon_{\infty,1})}\right] \quad (8)$$

where k is Boltzmann'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, and the indices 1 and 2 correspond to the polar solute and to the nonpolar solvent. As the dipole moments of *o*-xylene and *m*-xylene are much lower than the dipole moment of propylene carbonate (0.54 D, 0.35 D, and 4.98 D, correspondingly; at 25 °C, McClelan, 1989), we can calculate the apparent dipole moment of propylene carbonate, approximately, from the above equation. There are no measurements for concentrations lower than $x_1 = 0.1$, so we are not able to estimate the behavior of the system in this region. The variation of $\langle\mu\rangle$, at 25 °C, with the mole fraction of propylene carbonate is presented in Figure 6. Both systems show a change in the slope at $x_1 \approx 0.2$, which is more clear for the propylene carbonate + *m*-xylene system. Similar behavior has been observed in mixtures of *o*-xylene and *m*-xylene with γ-butyrolactone (Moumouzias and Ritzoulis, in press) and in other systems as well (Stokes and Marsh, 1976; French et al., 1988).

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