Physical Properties of the Ternary Mixture Dimethyl Carbonate + Methanol + Benzene and Its Corresponding Binaries at 298.15 K

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Density, refractive index, and speed of sound of the binary dimethyl carbonate + methanol, dimethyl carbonate + benzene and methanol + benzene and the ternary dimethyl carbonate + methanol + benzene mixtures have been measured at 298.15 K and atmospheric pressure, over the entire composition range. These results are used to calculate excess molar volumes, changes of refractive index on mixing, and deviations in isentropic compressibility for the above systems. The calculated quantities are further fitted to the Redlich−Kister and the Cibulka equations to estimate the binary and ternary fitting parameters, respectively, and root-mean-square deviations from the regression lines. Values of derived and excess properties were estimated and compared by different methods. Excess partial molar volumes at infinite dilution are also calculated.

1. Introduction

The thermodynamic study of esters of carbonic acid is arousing an increasing interest due to their uses as raw material in synthetic resins such as polycarbonate, in the synthesis of pharmaceuticals, and in agricultural chemistry. A method for manufacturing carbonic acid esters by transesterifying dimethyl carbonate with a hydroxy compound in the presence of a transesterification catalyst is improved by using benzene as an agent to make an azeotropic mixture with methanol and removing it azeotropically with benzene. This transesterification reaction is usually an equilibrium reaction, and it is necessary for success to remove the methanol or carbonic acid esters produced in order to sustain the reaction. Knowledge of the physical and derived properties of azeotropic liquid mixtures is required for the engineering design of extractive rectification columns, which are related to industrial separation processes.

In this work the densities, refractive indices, and speeds of sound of homogeneous binary and ternary mixtures at 298.15 K and atmospheric pressure have been measured. The results were used to calculate excess molar volumes, changes of refractive index on mixing, and deviations in isentropic compressibility over the entire mole fraction range for the mixtures. Experimental values of binary mixtures were correlated by the Redlich−Kister (1948) equation. The binary contribution was used to correlate the experimental ternary results with the Cibulka (1982) equation. The root-mean-square deviations are shown. The obtained values of excess molar volumes, changes of refractive index on mixing, and deviations in isentropic compressibility were compared with the results obtained by applying several equations (Iglesias et al., 1996) that predict excess and derived properties from the Redlich−Kister fitting parameters of binary systems. Their root-mean-square deviations are shown. By means of the calculated Redlich−Kister correlation parameters, the partial excess molar volumes corresponding to limiting values are being evaluated as a function of mole fraction.

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Table 1. Comparison of Data with Literature Data for Pure Liquids at 298.15 K

	ρ/(g·cm^{-3})		n_D	
component	exptl	lit.	exptl	lit.
dimethyl carbonate	1.0635	1.06350[a]	1.36640	1.36670[a]
		1.06320[b]		1.36670[b]
methanol	0.7866	0.78664[c]	1.32645	1.32652[c]
		0.78664[d]		1.32652[d]
benzene	0.8736	0.87366[c]	1.49729	1.49790[a]
		0.87366[d]		1.49792[c]
		0.87360[e]		1.49794[d]

[a] García et al. (1992). [b] Pal et al. (1998). [c] Das et al. (1994). [d] Riddick et al. (1986). [e] Kurihara et al. (1998).

2. Experimental Section

The chemicals were supplied by Merck except dimethyl carbonate by Fluka. The pure components were degassed ultrasonically and dried over molecular sieves Type 3 Å and 4 Å (supplied by Aldrich) and kept in an inert argon atmosphere as soon as the bottles were opened. Chromatographic (GLC) tests of the solvents showed purities which fulfilled purchaser specifications. Their mass fraction purities were more than 99 mass % for dimethyl carbonate and more than 99.8 mass % for methanol and benzene.. The maximum water contents of the liquids were determined using a Metrohm 737 KF coulometer. The corresponding obtained values were 1.5×10^{-2} for methanol, 1.1×10^{-2} for dimethyl carbonate, and 6×10^{-3} % for benzene. The solvent purities were compared with recent published values. The results are listed in Table 1.

The mixtures were prepared by weighing amounts of the pure liquids by syringing into stoppered bottles to prevent evaporation and reducing possible errors in mole fraction calculations. A Mettler AT-261 Delta Range balance was used with a precision of $\pm 10^{-5}$ g, covering the whole composition range of the mixture. The density and the speed of sound of the pure liquids and mixtures were

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Table 2. Density ρ, Refractive Index n_D, Excess Molar Volume V_m^E, Change of Refractive Index on Mixing Δn_D, Speed of Sound u, Isentropic Compressibility κ_S, and Deviation in Isentropic Compressibility $\Delta\kappa_S$ for Binary Mixtures at 298.15 K

x_1	ρ/(g·cm^{-3})	n_D	u/(m·s^{-1})	V_m^E/(cm^3·mol^{-1})	Δn_D	κ_S/(T·Pa^{-1})	$\Delta\kappa_S$/(T·Pa^{-1})
Dimethyl Carbonate (1) + Methanol (2)							
0	0.7866	1.32645	1102	0	0	1046.6	0
0.0506	0.8144	1.33069	1109	−0.009	0.0022	997.5	−29.4
0.1042	0.8409	1.33465	1117	−0.020	0.0040	953.9	−52.0
0.2079	0.8851	1.34105	1129	−0.041	0.0063	886.0	−79.6
0.3011	0.9184	1.34593	1140	−0.056	0.0074	838.2	−91.0
0.3963	0.9475	1.35025	1149	−0.066	0.0080	798.7	−93.4
0.4931	0.9729	1.35390	1159	−0.065	0.0077	765.2	−89.2
0.5890	0.9948	1.35692	1168	−0.059	0.0069	736.4	−80.6
0.6927	1.0155	1.35993	1177	−0.049	0.0058	711.0	−65.6
0.8137	1.0365	1.36284	1187	−0.034	0.0039	684.9	−44.5
0.8997	1.0497	1.36460	1193	−0.022	0.0022	669.0	−26.9
0.9575	1.0579	1.36567	1196	−0.015	0.0010	661.2	−12.2
1	1.0635	1.36640	1196	0	0	656.8	0
Dimethyl Carbonate (1) + Benzene (2)							
0	0.8736	1.49729	1299	0	0	678.2	0
0.0528	0.8825	1.48985	1292	0.063	−0.0005	678.5	1.5
0.1026	0.8910	1.48310	1285	0.115	−0.0008	679.3	3.3
0.2044	0.9087	1.46936	1271	0.201	−0.0012	680.6	6.8
0.3038	0.9264	1.45615	1259	0.260	−0.0014	681.4	9.7
0.4054	0.9449	1.44277	1246	0.298	−0.0015	681.2	11.7
0.5404	0.9702	1.42515	1232	0.310	−0.0014	679.5	12.8
0.5999	0.9816	1.41743	1226	0.303	−0.0013	678.0	12.6
0.6944	1.0000	1.40524	1218	0.281	−0.0012	674.5	11.2
0.7881	1.0188	1.39333	1210	0.228	−0.0008	670.4	9.0
0.8806	1.0379	1.38151	1204	0.150	−0.0005	665.1	5.8
0.9360	1.0497	1.37454	1200	0.082	−0.0002	661.2	3.0
Methanol (1) + Benzene (2)							
0.0745	0.8703	1.49099	1282	0.022	0.0064	699.2	−6.4
0.1756	0.8656	1.48146	1263	0.028	0.0142	724.5	−18.4
0.2762	0.8604	1.47113	1248	0.028	0.0210	746.4	−33.6
0.3885	0.8538	1.45802	1232	0.022	0.0271	772.2	−49.1
0.4832	0.8475	1.44542	1217	0.009	0.0307	796.5	−59.7
0.5912	0.8391	1.42826	1200	−0.004	0.0320	827.2	−68.8
0.6871	0.8303	1.41072	1183	−0.014	0.0308	860.6	−70.7
0.8247	0.8145	1.37945	1153	−0.013	0.0230	923.3	−58.7
0.8909	0.8052	1.36167	1136	−0.010	0.0166	962.6	−43.8
0.9629	0.7934	1.33903	1115	−0.001	0.0062	1014.1	−18.8

measured with an Anton Paar DSA-48 densimeter and sound analyzer with a precision of $\pm10^{-4}$ g·cm^{-3} and ±1 m·s^{-1} The refractive index was measured with the automatic refractometer ABBEMAT-HP Dr Kernchen with a precision of $\pm10.^{-5}$ Before these kinds of measurements, these instruments were calibrated with Millipore quality water and ambient air, respectively, in accordance with the instructions.

3. Results and Discussion

Density, refractive index, speed of sound, excess molar volume, change of refractive index on mixing, isentropic compressibility (determined by means of the Laplace equation, $\kappa_S = \rho^{-1}\cdot u^{-2}$), and deviation in isentropic compressibility $\Delta\kappa_S$ of the binary and ternary mixtures are reported in Tables 2 and 3, respectively. Excess molar volumes, changes of refractive indices on mixing, and deviations in isentropic compressibility for binary and ternary mixtures were derived, respectively, from eqs 1, 2, and 3.

$$V_m^E = \sum_{i=1}^{N} x_i\cdot M_i\cdot(\rho^{-1} - \rho^{\circ -1}) \quad (1)$$

$$\Delta n_D = n_D - \sum_{i=1}^{N} x_i n_{Di}^{\circ} \quad (2)$$

$$\Delta\kappa_S = \kappa_S - \sum_{i=1}^{N} x_i\kappa_{S,i} \quad (3)$$

In these equations, ρ and n_D are the density and refractive index of the mixture, ρ_i° and n_{Di}° are the density and refractive index of the pure components, κ_S is the isentropic compressibility of the mixture, and $\kappa_{S,i}$ is the isentropic compressibility of the pure component.

The binary values were fitted to a Redlich−Kister type equation:

$$\Delta Q = x_i\cdot x_j\cdot\sum_{p=0}^{m} B_p\cdot(x_i - x_i)^p \quad (4)$$

where ΔQ_{ij} is the excess property, x is the mole fraction, B_P is the fitting parameter, and M is the degree of the polynomic expansion. Ternary results were correlated with the Cibulka equation.

$$\Delta Q_{123} = \Delta Q_{12} + \Delta Q_{13} + \Delta Q_{23} + x_1x_2(1 - x_1 - x_2)(C_1 + C_2\cdot x_1 + C_3\cdot x_2) \quad (5)$$

where ΔQ_{12}, ΔQ_{13} and ΔQ_{23} are the parameters representing the binary interactions in accordance with the Redlich−Kister expression. The degree of this equation was optimized by applying the F-test (Bevington, 1969). The correlation parameters calculated using eqs 4 and 5 are listed in Table 4, together with the root-mean-square

Table 3. Density ρ, Refractive Index n_D, Excess Molar Volume V_m^E, Change of Refractive Index on Mixing Δn_D, Speed of Sound u, Isentropic Compressibility κ_S, and Deviation in Isentropic Compressibility $\Delta\kappa_S$ for a Ternary Mixture at 298.15 K

x_1	x_2	ρ/(g·cm^{-3})	n_D	u/(m·s^{-1})	V_m^E/(cm^3·mol^{-1})	Δn_D	κ_S/(T·Pa^{-1})	$\Delta\kappa_S$/(T·Pa^{-1})
Dimethyl Carbonate (1) + Methanol (2) + Benzene (3)								
0.0490	0.9094	0.8197	1.34389	1121	−0.017	0.0084	969.9	−42.2
0.1092	0.7925	0.8528	1.36302	1143	−0.006	0.0154	896.9	−71.0
0.2699	0.2748	0.9167	1.42945	1219	0.158	0.0144	734.5	−39.1
0.0933	0.6837	0.8569	1.39200	1170	0.009	0.0237	852.8	−75.2
0.2841	0.5997	0.9150	1.37214	1162	0.007	0.0145	809.5	−83.5
0.2068	0.5694	0.8945	1.39283	1177	0.044	0.0199	807.3	−76.2
0.1027	0.5760	0.8668	1.41121	1189	0.028	0.0258	816.0	−72.2
0.3937	0.4845	0.9453	1.37565	1171	0.030	0.0127	770.8	−77.4
0.2884	0.4919	0.9180	1.39254	1181	0.053	0.0170	780.9	−72.3
0.1823	0.4978	0.8909	1.41019	1192	0.062	0.0218	789.5	−68.2
0.0934	0.5006	0.8688	1.42573	1203	0.044	0.0262	795.4	−65.2
0.4769	0.4042	0.9654	1.37642	1177	0.044	0.0106	747.3	−69.6
0.3818	0.4012	0.9416	1.39263	1187	0.095	0.0138	754.2	−63.6
0.2868	0.3988	0.9187	1.40852	1197	0.113	0.0169	759.9	−59.1
0.1897	0.4044	0.8959	1.42399	1207	0.093	0.0206	766.0	−57.1
0.1018	0.3839	0.8767	1.44167	1221	0.067	0.0233	765.3	−52.1
0.5694	0.3019	0.9849	1.37953	1186	0.072	0.0083	721.6	−55.6
0.4727	0.3024	0.9621	1.39417	1194	0.123	0.0104	728.9	−50.5
0.3718	0.3087	0.9389	1.40886	1203	0.152	0.0130	736.1	−47.9
0.2869	0.3056	0.9200	1.42265	1212	0.148	0.0151	739.9	−44.7
0.1883	0.3099	0.8985	1.43756	1222	0.113	0.0179	745.2	−43.1
0.1245	0.2615	0.8865	1.45360	1237	0.106	0.0173	737.3	−34.5
0.7193	0.1413	1.0119	1.38289	1198	0.122	0.0039	687.7	−27.2
0.5827	0.1851	0.9836	1.39566	1203	0.177	0.0063	702.5	−31.4
0.4871	0.1886	0.9631	1.40877	1211	0.198	0.0075	708.5	−28.7
0.4027	0.1860	0.9453	1.42138	1219	0.203	0.0086	711.9	−26.2
0.2950	0.1906	0.9231	1.43650	1229	0.186	0.0104	716.8	−25.3
0.1930	0.1963	0.9023	1.45071	1240	0.158	0.0122	720.8	−25.5
0.0951	0.1851	0.8834	1.46662	1252	0.102	0.0134	722.2	−22.1
0.7735	0.0971	1.0212	1.38197	1201	0.133	0.0025	679.0	−18.4
0.6725	0.1012	0.9997	1.39501	1208	0.204	0.0030	685.6	−15.5
0.5721	0.0996	0.9788	1.40875	1217	0.241	0.0034	690.2	−12.5
0.4839	0.0970	0.9608	1.42123	1225	0.264	0.0038	693.0	−10.6
0.3836	0.1017	0.9410	1.43441	1235	0.254	0.0047	696.7	−10.7
0.2889	0.1028	0.9227	1.44761	1245	0.222	0.0057	699.0	−10.8
0.1895	0.1049	0.9037	1.46133	1256	0.183	0.0068	701.8	−10.9
0.0935	0.1013	0.8860	1.47526	1267	0.116	0.0075	703.3	−10.2
0.8739	0.0589	1.0406	1.37415	1199	0.079	0.0013	668.9	−12.3
0.0484	0.0468	0.8799	1.48648	1283	0.074	0.0035	690.6	−3.8
0.1955	0.6780	0.8869	1.37233	1157	0.004	0.0165	842.6	−81.2
0.6748	0.1985	1.0049	1.38049	1194	0.096	0.0054	698.3	−38.6

Table 4. Parameters and Root-Mean-Square Deviations σ

Dimethyl Carbonate (1) + Methanol (2)					
V^E_m/(cm^3·mol^{-1})	$B_0 = -0.2514$	$B_1 = 0.0195$			$\sigma = 0.003$
Δn_D	$B_0 = 0.0306$	$B_1 = -0.0110$	$B_2 = 0.0047$		$\sigma = 0.00006$
$\Delta\kappa_S$/(T·Pa^{-1})	$B_0 = -353.7671$	$B_1 = 160.4949$	$B_2 = -112.7878$		$\sigma = 0.6$
Dimethyl Carbonate (1) + Benzene (2)					
V^E_m/(cm^3·mol^{-1})	$B_0 = 1.2468$	$B_1 = 0.1058$	$B_2 = 0.1508$		$\sigma = 0.002$
Δn_D	$B_0 = -0.0058$	$B_1 = 0.0010$	$B_2 = -0.0012$	$B_3 = -0.0025$	$\sigma = 0.00003$
$\Delta\kappa_S$/(T·Pa^{-1})	$B_0 = 50.9165$	$B_1 = 10.3352$	$B_2 = -9.6475$		$\sigma = 0.1$
Methanol (1) + Benzene (2)					
V^E_m/(cm^3·mol^{-1})	$B_0 = 0.0240$	$B_1 = -0.2355$	$B_2 = 0.0865$		$\sigma = 0.002$
Δn_D	$B_0 = 0.1239$	$B_1 = 0.0473$	$B_2 = 0.0125$		$\sigma = 0.00011$
$\Delta\kappa_S$/(T·Pa^{-1})	$B_0 = -246.9458$	$B_1 = -191.1843$	$B_2 = -51.1417$		$\sigma = 0.3$
Dimethyl Carbonate (1) + Methanol (2) + Benzene (3)					
V^E_m/(cm^3·mol^{-1})	$C_1 = -0.3544$	$C_2 = 1.6341$	$C_3 = 1.2202$		$\sigma = 0.004$
Δn_D	$C_1 = -0.0091$	$C_2 = -0.0317$	$C_3 = -0.1065$		$\sigma = 0.00008$
$\Delta\kappa_S$/(T·Pa^{-1})	$C_1 = -75.5943$	$C_2 = 156.7012$	$C_3 = 919.1128$		$\sigma = 0.4$

deviations (σ). This deviation is calculated by applying the following expression:

$$\sigma = \left(\frac{\sum_i^{n_{DAT}} (z_{exp} - z_{pred})^2}{n_{DAT}} \right)^{1/2} \qquad (6)$$

where property values and the number of experimental data are represented by z and n_{DAT}, respectively.

The fitted curves, as well as excess changes on mixing and deviation values at 298.15 K for binary mixtures, are shown in Figures 1−3 together with comparisons with literature data. Excess molar volumes change sign between mixtures containing benzene and methanol. For the binary mixture methanol + benzene the fitted curve of excess molar volumes is positive at high mole fraction of methanol followed by a negative lobe at approximately equimolar composition. For changes of refractive index on mixing and deviations in isentropic compressibility the behavior is

Table 5. Root-Mean-Square Deviations of the Experimental Results from the Prediction Results for Several Empirical Equations

	$\sigma(V_m^E/(cm^3 \cdot mol^{-1}))$	$\sigma(\Delta n_D)$	$\sigma(\Delta\kappa_S/(T \cdot Pa^{-1})$
Kohler	0.014	0.0015	9.3
Jacob-Fitzner	0.015	0.0013	7.1
Colinet	0.013	0.0014	8.3
Tsao-Smith[a]	0.017	0.0064	22.1
Tsao-Smith[b]	0.042	0.0002	2.8
Tsao-Smith[c]	0.025	0.0037	26.7
Scatchard[a]	0.017	0.0017	12.8
Scatchard[b]	0.009	0.0017	1.6
Scatchard[c]	0.016	0.0026	13.0
Toop[a]	0.017	0.0015	12.1
Toop[b]	0.010	0.0002	1.6
Toop[c]	0.016	0.0025	12.1

[a] Benzene is the asymmetric component in the equation. [b] Methanol is the asymmetric component in the equation. [c] Dimethyl carbonate is the asymmetric component in the equation.

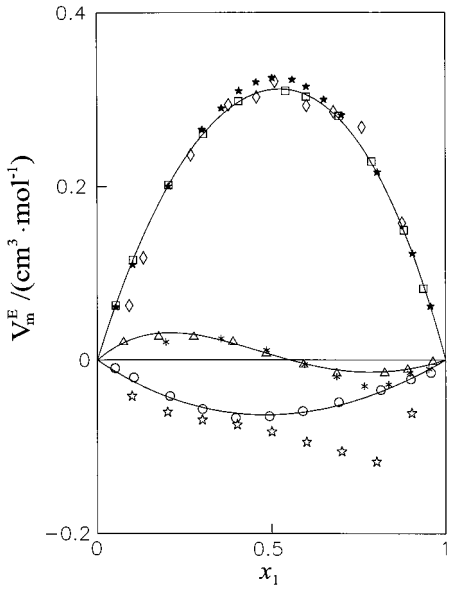

Figure 1. Curves of excess molar volumes $V_m^E/(cm^3 \cdot mol^{-1})$ from the Redlich−Kister equation (eq 4) at 298.15 K for (○) dimethyl carbonate (1) + methanol (2), **(△)** methanol (1) + benzene (2) and (□) dimethyl carbonate (1) + benzene (2), experimental values and fitted curves. Comparison with literature data: (☆) dimethyl carbonate (1) + methanol (2) (Aminabhavi et al, 1998); (*) methanol (1) + benzene (2) (Miyano et al, 1998); (◊) (Negadi et al, 1993); (★) dimethyl carbonate(1) + benzene (2) (García de la Fuente et al., 1992).

opposite for the binary mixture dimethyl carbonate + benzene: it is negative and positive, respectively, for the corresponding derived properties. The same occurs for the other binaries. The speed of sound, isentropic compressibility, and excess molar volume are the properties sensitive to different kinds of association in the pure components and in the mixtures, and they are often related to local order. These data will be used in future papers after studying these properties at different temperatures, which will give us information about the molecular packing, various types of intermolecular interactions, and their strengths, all this influenced by the size, shape, and chemical nature of component molecules. We could investigate in this way the tendency of interstitial accommodation of methanol molecules into the other components, dimethyl carbonate and benzene, leading to negative excess molar volumes.

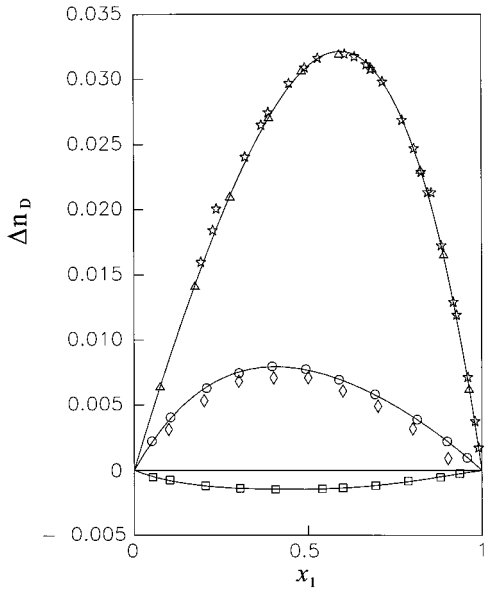

Figure 2. Curves of changes of refractive index on mixing Δn_D from the Redlich−Kister equation (eq 4) at 298.15 K for (○) dimethyl carbonate (1) + methanol (2), **(△)** methanol (1) + benzene (2), and (□) dimethyl carbonate (1) + benzene (2). Comparison with literature data: (☆) methanol (1) + benzene (2) (Arce et al, 1980) and (◊) dimethyl carbonate (1) + methanol (2) (Aminabhavi et al, 1998).

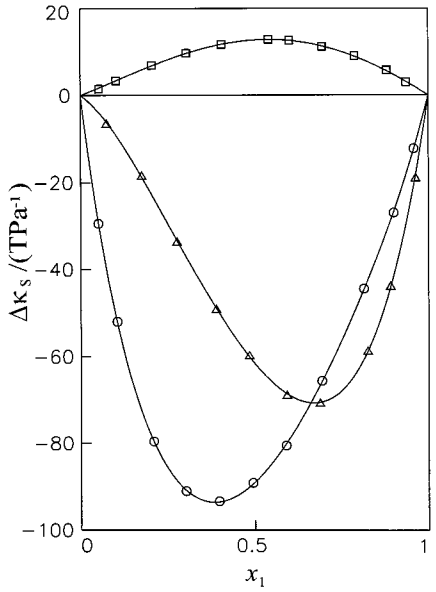

Figure 3. Curves of deviations in isentropic compressibility $\Delta K_s/(T \cdot Pa^{-1})$ from the Redlich−Kister equation (eq 4) at 298.15 K for (○) dimethyl carbonate (1) + methanol (2), **(△)** methanol (1) + benzene (2), and (□) dimethyl carbonate (1) + benzene (2).

Figures 4−6 exhibit the derived properties for the ternary mixture dimethyl carbonate + methanol + benzene. Excess molar volumes are positive over most of the composition diagram, except when they are close to pure methanol, where a change in sign occurs. Changes of refractive index on mixing are positive. Deviations in isentropic compressibility reproduce the same behavior as that for excess molar volumes for binary mixtures. Values are positive, when they are close to the binary mixture dimethyl carbonate + benzene.

Some predictive methods for ternary excess properties are based on additive binary contributions. In Table 5 the

Table 6. Partial Excess Molar Volumes at Infinite Dilution of the Binary Mixtures at 298.15 K

system	$V_1^{E,\infty}$/(cm^3·mol^{-1})	$V_2^{E,\infty}$/(cm^3·mol^{-1})
dimethyl carbonate (1) + methanol (2)	−0.271	−0.232
dimethyl carbonate (1) + benzene (2)	1.292	1.503
methanol (1) + benzene (2)	0.346	−0.125

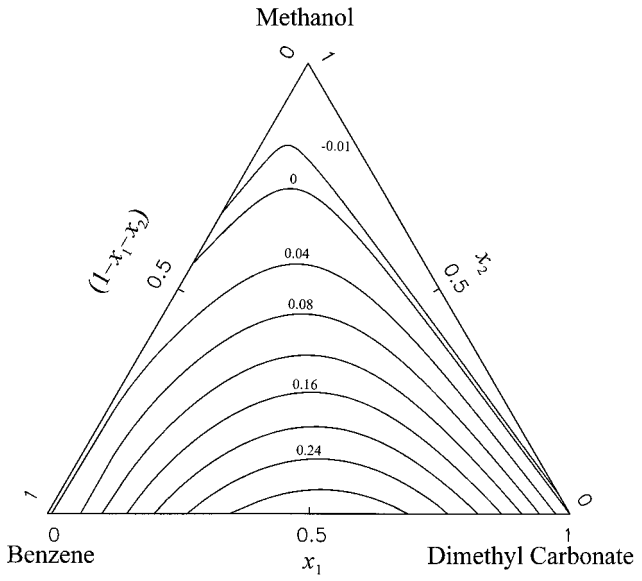

Figure 4. Curves of constant excess molar volumes V^E_m/(cm^3·mol^{-1}) from the Cibulka equation (eq 5) at 298.15 K for dimethyl carbonate (1) + methanol (2) + benzene (3).

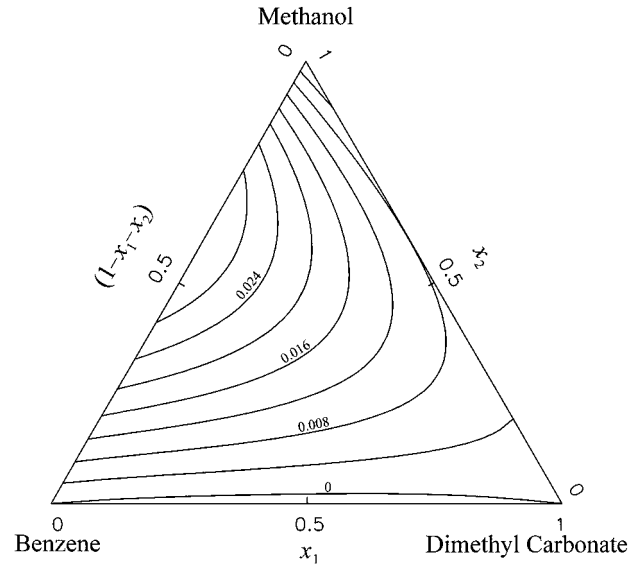

Figure 5. Curves of constant changes of refractive index on mixing Δn_D from the Cibulka equation (eq 5) at 298.15 K for dimethyl carbonate (1) + methanol (2) + benzene (3).

experimental and estimated excess values are compared and corresponding root-mean-square deviations are shown. In general for the ternary system the best estimations of excess molar volumes are given when applying symmetric equations. For changes of refractive index on mixing and deviations in isentropic compressibility, symmetric equations are the best; however, asymmetric equations give good predictions when a type b equation is used.

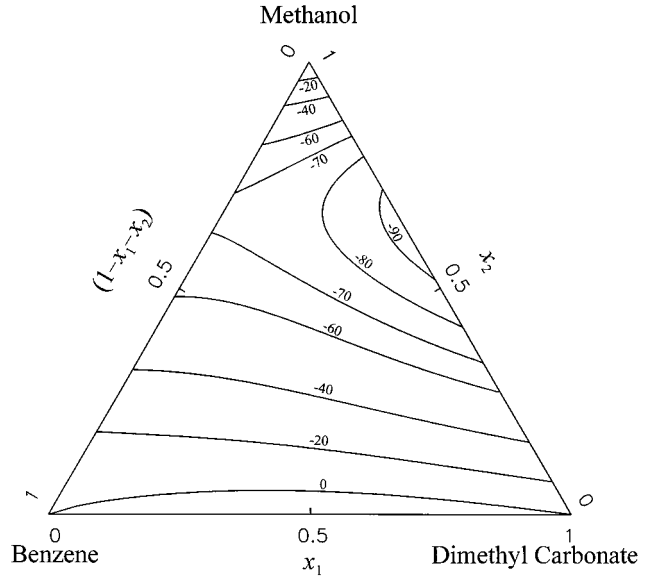

Figure 6. Curves of constant deviations in isentropic compressibility $\Delta\kappa_S$/(T·Pa^{-1}) from the Cibulka equation (eq 5) at 298.15 K for dimethyl carbonate (1) + methanol (2) + benzene (3).

In Table 6, values of limiting pure partial excess molar volumes (Canosa et al., 1997) at 298.15 K for the binary mixtures are shown.

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Registry Nos. Supplied by the Author: Dimethyl carbonate, 616-38-6; methanol, 67-56-1; benzene, 71-43-2.

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