

Liquid–Liquid Equilibrium and Physical Properties of the Ternary Mixture (Dimethyl Carbonate + Methanol + Cyclohexane) at 298.15 K

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Density, refractive index, and speed of sound of dimethyl carbonate (1) + cyclohexane (3) and methanol (2) + cyclohexane (3) and dimethyl carbonate (1) + methanol (2) + cyclohexane (3) have been measured at 298.15 K and atmospheric pressure, over the miscible composition range. Excess molar volumes, changes of refractive index on mixing, and deviations in isentropic compressibility for the systems have been calculated. Redlich–Kister and Cibulka equations have been used to estimate the binary and ternary fitting parameters, and root-mean-square deviations from the regression lines are shown. Values of derived and excess properties were estimated by different methods and compared with experimental data. Excess partial molar volumes at infinite dilution are also calculated. The liquid–liquid equilibrium of dimethyl carbonate (1) + methanol (2) + cyclohexane (3) at 298.15 K and atmospheric pressure has been measured. The UNIQUAC equation was used to correlate the experimental data, and the root-mean-square deviation between experimental and predictive values is shown.

1. Introduction

Densities, refractive indices, and speeds of sound in the miscible region of binary and ternary mixtures of dimethyl carbonate (1) + methanol (2) + cyclohexane (3) at 298.15 K and atmospheric pressure have been measured. This is a continuation of the thermodynamic study¹ of the multi-component mixture containing dimethyl carbonate and methanol due to the increasing interest in carbonic acid as a raw material in synthetic resins such as polycarbonate, in the synthesis of pharmaceuticals, and in agricultural chemistry. In previous papers,¹ the density, refractive index, and speed of sound of the binary mixture dimethyl carbonate (1) + methanol (2) were measured.

The experimental values were used to calculate excess molar volumes, changes of refractive index on mixing, and deviations in isentropic compressibility over the miscible mole fraction range for the mixtures. Experimental values were correlated with Redlich–Kister² and Cibulka³ equations for binary and ternary mixtures, respectively. The root-mean-square deviation between experimental and calculated values is shown. Comparison between experimental and literature data has been made for binary mixtures.^{4–6} The excess molar volumes, changes of refractive index on mixing, and deviations in isentropic compressibility were compared with the results obtained by applying several equations⁷ 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 values⁷ have been evaluated.

The liquid–liquid equilibrium of dimethyl carbonate (1) + methanol (2) + cyclohexane (3) has been measured because a region of immiscibility was presented when the physical properties were studied. The experimental tie lines were used to obtain, by applying the UNIQUAC equation,⁸ the binodal curve. Experimental data of the liquid–liquid

Table 1. Comparison of Density ρ and Refractive Index n_D with Literature Data for Pure Components at 298.15 K

component	ρ (g·cm ⁻³)		n_D	
	exptl	lit.	exptl	lit.
dimethyl carbonate	1.0635	1.063 50 ^a 1.063 20 ^b	1.366 40	1.366 70 ^{a,b}
methanol	0.7866	0.786 64 ^{c,d}	1.326 45	1.326 52 ^{c,d}
cyclohexane	0.7738	0.773 89 ^d	1.423 63	1.423 54 ^d

^a García et al.⁵ ^b Pal et al.¹⁰ ^c Das et al.¹¹ ^d Riddick et al.¹²

equilibrium related with the ternary system were not found in the literature data.

2. Experimental Section

The pure components were supplied by Merck. Dimethyl carbonate was supplied by Fluka. Their mass fraction purities were >99 mass % for dimethyl carbonate, >99.8 mass % for methanol, and >99.5 mass % for cyclohexane. Materials were degassed ultrasonically, dried over molecular sieves Type 3 Å and 4 Å (Aldrich), and kept in an inert argon atmosphere (with a maximum content in water of 2.14×10^{-6} by mass fraction). Chromatographic analysis of the solvents showed purities which fulfilled purchaser specifications. The maximum water contents of the liquids were determined using a Metrohm 737 KF coulometer. The corresponding obtained values were 1.1×10^{-2} mass % for methanol and negligible quantities for the rest of the liquids. The densities and refractive indices of the components were compared with recently published values in Table 1.

The mixtures were prepared from known masses of the pure liquids, by syringing into stoppered bottles to prevent evaporation and reduce possible errors in the mole fractions. 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 measured with an

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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	$\frac{\rho}{\text{g}\cdot\text{cm}^{-3}}$	n_D	$\frac{u}{\text{m}\cdot\text{s}^{-1}}$	$\frac{V_m^E}{\text{cm}^3\cdot\text{mol}^{-1}}$	Δn_D	$\frac{\kappa_S}{\text{T}\cdot\text{Pa}^{-1}}$	$\frac{\Delta\kappa_S}{\text{T}\cdot\text{Pa}^{-1}}$
Dimethyl Carbonate (1) + Cyclohexane (2)							
0	0.7738	1.423 63	1255	0	0		0
0.0378	0.7801	1.420 71	1244	0.318	-0.0008	829	14
0.0808	0.7879	1.417 41	1232	0.605	-0.0016	835	28
0.1764	0.8070	1.410 74	1212	1.064	-0.0028	843	51
0.2896	0.8325	1.403 53	1195	1.358	-0.0035	841	68
0.3880	0.8570	1.397 79	1184	1.457	-0.0036	832	75
0.4938	0.8856	1.391 87	1176	1.451	-0.0035	817	77
0.5908	0.9141	1.386 54	1172	1.348	-0.0033	796	72
0.6864	0.9443	1.381 47	1172	1.180	-0.0029	770	62
0.7918	0.9807	1.376 12	1176	0.885	-0.0022	737	46
0.8976	1.0210	1.371 11	1185	0.468	-0.0011	698	24
0.9503	1.0423	1.368 64	1190	0.245	-0.0006	677	12
1	1.0635	1.366 40	1196	0	0	657	0
Methanol (2)+ Cyclohexane (3)							
0	0.7738	1.423 63	1255	0	0	821	0
0.0198	0.7733	1.422 68	1248	0.083	0.0010	830	4
0.0286	0.7731	1.422 41	1247	0.116	0.0016	832	5
0.0484	0.7729	1.421 46	1242	0.155	0.0025	839	8
0.0550	0.7728	1.421 12	1240	0.173	0.0028	841	8
0.8309	0.7745	1.355 87	1113	0.512	0.0130	1041	33
0.8960	0.7773	1.345 48	1105	0.386	0.0089	1054	31
0.9566	0.7819	1.334 66	1101	0.185	0.0040	1055	18
1	0.7866	1.326 45	1102	0	0	1047	0

Anton Paar DSA-48 densimeter and sound analyzer with a precision of $\pm 10^{-4}$ g·cm⁻³ and ± 1 m·s⁻¹. The refractive index at the sodium D line was measured by the automatic refractometer ABBEMAT-HP Dr Kernchen with a precision of $\pm 10^{-5}$. These instruments were calibrated with Millipore quality water and ambient air, respectively, in accordance with the instructions.

The apparatus for the liquid–liquid equilibrium determination consists of a glass cell with a water jacket in order to maintain a constant temperature. Temperature was controlled within $\pm 5 \times 10^{-2}$ K inside the cell. The cell was connected to a controller bath Polyscience model 9010, with a stability of $\pm 10^{-2}$ K. The temperature in the cell was measured with an Anton Paar MKT-100 digital thermometer (precision $\pm 10^{-2}$ K) over the entire range of temperatures. The estimated uncertainty in the mole fraction for the equilibrium data was 3×10^{-3} in the cyclohexane rich region (phase I) and 2×10^{-3} in the methanol rich region (phase II).

3. Results and Discussion

Density, refractive index, speed of sound, excess molar volume, changes of refractive index on mixing, isentropic compressibility (determined by means of the Laplace equation, $\kappa_S = \rho^{-1}u^{-2}$, where u is the speed of sound), and deviation in isentropic compressibility of the miscibility region for 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

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

$$\Delta n_D = n_D - \sum_{i=1}^N x_i n_{D,i}^0 \quad (2)$$

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

In these equations, ρ and n_D are the density and refractive index of the mixture, ρ_i^0 and $n_{D,i}^0$ are the densities and refractive indexes of the pure components, κ_S is the isentropic compressibility of the mixture, and $\kappa_{S,i}$ are the isentropic compressibilities of the pure components.

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

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

where ΔX_{ij} is the excess property, x is the mole fraction, B_p is the fitting parameter, and m is the degree of the polynomial expansion. For ternary mixtures the corresponding equation is

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

where ΔX_{12} , ΔX_{13} , and ΔX_{23} are the parameters representing the binary interactions in accordance with the Redlich–Kister expression and C_1 , C_2 , and C_3 are the fitting parameters. Applying the F-test,⁹ the degree of the polynomial expression was optimized. The correlation parameters calculated using eqs 4 and 5 are listed in Table 4, together with the root-mean-square deviation σ . This deviation is calculated by applying the following expression:

$$\sigma = \left(\sum_i^{n_{\text{DAT}}} (z_{\text{exp}} - z_{\text{calc}})^2 / n_{\text{DAT}} \right)^{1/2} \quad (6)$$

where experimental and calculated data are represented by z_{exp} and z_{calc} , respectively, and the number of experimental data is represented by n_{DAT} .

Figure 1 shows the excess molar volumes; they are positive in the entire composition range. Dimethyl carbonate (1) + cyclohexane (3) are miscible over the whole composition range.

Figure 2 exhibits the excess molar volumes for dimethyl carbonate (1) + methanol (2) + cyclohexane (3). Excess molar volumes are positive over most of the composition

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 Ternary Mixtures 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) + Cyclohexane (3)								
0.0587	0.0624	0.7833	1.416 12	1220	0.630	0.0019	858	32
0.0471	0.9072	0.8060	1.338 37	1106	0.186	0.0056	1015	-3
0.1011	0.0923	0.7920	1.411 42	1202	0.894	0.0025	873	48
0.0849	0.1946	0.7912	1.407 33	1190	0.705	0.0075	893	42
0.1129	0.2993	0.8011	1.398 79	1170	0.733	0.0107	912	42
0.1106	0.3839	0.8035	1.393 02	1160	0.670	0.0130	924	35
0.1153	0.4527	0.8072	1.387 17	1150	0.686	0.0141	937	33
0.1087	0.5858	0.8102	1.375 48	1133	0.691	0.0150	961	26
0.1074	0.7004	0.8167	1.362 54	1119	0.599	0.0131	978	17
0.0973	0.7967	0.8212	1.350 59	1111	0.399	0.0100	986	2
0.1980	0.0938	0.8136	1.404 69	1184	1.256	0.0015	876	67
0.2075	0.1865	0.8201	1.398 61	1167	1.185	0.0050	895	66
0.1996	0.3039	0.8245	1.391 77	1153	1.011	0.0091	912	55
0.2722	0.2299	0.8405	1.391 56	1157	1.219	0.0058	889	61
0.1135	0.6222	0.8140	1.371 08	1127	0.674	0.0144	967	25
0.1690	0.6329	0.8379	1.364 86	1123	0.627	0.0124	946	10
0.2069	0.6798	0.8620	1.355 79	1121	0.413	0.0101	923	-18
0.3810	0.0711	0.8595	1.394 36	1173	1.472	-0.0006	846	72
0.2825	0.1885	0.8404	1.393 54	1161	1.301	0.0044	882	65
0.3080	0.2813	0.8557	1.385 61	1151	1.135	0.0069	882	48
0.2870	0.4064	0.8612	1.377 25	1139	0.892	0.0095	895	29
0.2884	0.4981	0.8728	1.368 93	1134	0.699	0.0102	892	6
0.3043	0.5909	0.8952	1.358 10	1132	0.380	0.0093	871	-33
0.3624	0.1026	0.8567	1.393 78	1169	1.437	0.0009	854	69
0.3941	0.1795	0.8732	1.386 98	1161	1.270	0.0033	850	53
0.3942	0.2934	0.8866	1.378 96	1152	0.991	0.0064	850	28
0.3512	0.4327	0.8895	1.370 44	1142	0.723	0.0090	863	2
0.3683	0.5129	0.9112	1.361 61	1141	0.409	0.0089	843	-34
0.4473	0.1105	0.8825	1.388 12	1167	1.357	0.0008	832	59
0.4853	0.1786	0.9031	1.381 64	1164	1.128	0.0031	817	35
0.4607	0.3101	0.9141	1.373 24	1156	0.790	0.0061	818	3
0.4807	0.4034	0.9406	1.364 22	1156	0.394	0.0073	796	-38
0.5854	0.0928	0.9249	1.381 32	1172	1.147	0.0002	787	41
0.5700	0.2038	0.9379	1.374 74	1170	0.822	0.0035	778	5
0.5711	0.3251	0.9637	1.365 37	1168	0.351	0.0060	761	-39
0.6655	0.1156	0.9574	1.375 44	1178	0.841	0.0011	752	14
0.6918	0.1999	0.9863	1.368 17	1181	0.400	0.0036	727	-26
0.7834	0.1108	1.0027	1.369 57	1187	0.424	0.0015	707	-10
0.8731	0.0723	1.0301	1.367 67	1191	0.224	0.0010	685	-9
0.1999	0.3816	0.8295	1.386 08	1146	0.916	0.0110	917	43
0.1973	0.5072	0.8376	1.375 28	1133	0.801	0.0122	930	27
0.2976	0.0907	0.8382	1.398 40	1174	1.436	0.0006	865	73
0.0637	0.7809	0.8026	1.356 32	1113	0.513	0.0122	1005	18

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

	Dimethyl Carbonate (1) + Cyclohexane (3)					
V_m^E /(cm ³ ·mol ⁻¹)	$B_0 = 5.7889$	$B_1 = -1.2193$	$B_2 = 1.2518$	$B_3 = -0.8412$		$\sigma = 0.006$
Δn_D	$B_0 = -0.0140$	$B_1 = 0.0041$	$B_2 = -0.0077$	$B_3 = 0.0014$	$B_4 = 0.0052$	$\sigma = 0.00003$
$\Delta\kappa_S$ /(T·Pa ⁻¹)	$B_0 = 305.43$	$B_1 = -44.95$	$B_2 = 20.65$	$B_3 = -33.35$		$\sigma = 0.2$
	Methanol (2) + Cyclohexane (3)					
V_m^E /(cm ³ ·mol ⁻¹)	$B_0 = 0.2942$	$B_1 = 3.9063$	$B_2 = 4.4812$	$B_3 = -4.1331$		$\sigma = 0.003$
Δn_D	$B_0 = 0.0764$	$B_1 = 0.0240$				$\sigma = 0.00004$
$\Delta\kappa_S$ /(T·Pa ⁻¹)	$B_0 = -47.36$	$B_1 = 145.72$	$B_2 = 423.98$			$\sigma = 0.2$
	Dimethyl Carbonate (1) + Methanol (2) + Cyclohexane (3)					
V_m^E /(cm ³ ·mol ⁻¹)	$C_1 = 29.394$	$C_2 = -33.696$	$C_3 = -26.133$			$\sigma = 0.005$
Δn_D	$C_1 = -0.0504$	$C_2 = 0.0220$	$C_3 = -0.0789$			$\sigma = 0.00007$
$\Delta\kappa_S$ /(T·Pa ⁻¹)	$C_1 = 2742.9$	$C_2 = -4304.5$	$C_3 = -379.9$			$\sigma = 0.6$

diagram, except when they are close to those of the binary mixture dimethyl carbonate (1) + methanol (2), where a change in sign occurs.

The liquid–liquid equilibrium for the ternary mixture has been determined at atmospheric pressure and 298.15 K. Table 5 gives the liquid–liquid composition of both phases. Figure 3 shows the experimental tie lines, the binodal curve obtained by applying the UNIQUAC equation, and the extrapolated plait point at 298.15 K.

The composition of this point is $x_1 = 0.086$ and $x_2 = 0.422$ for dimethyl carbonate and methanol, respectively. Table 6 shows a comparison between the experimental results and literature values of mutual solubility data for the binary methanol (2) + cyclohexane (3) mixture at 298.15 K. In Table 7 the correlated parameters for the UNIQUAC equation and the root-mean-square deviation from the results of the UNIQUAC model at 298.15 K are shown.

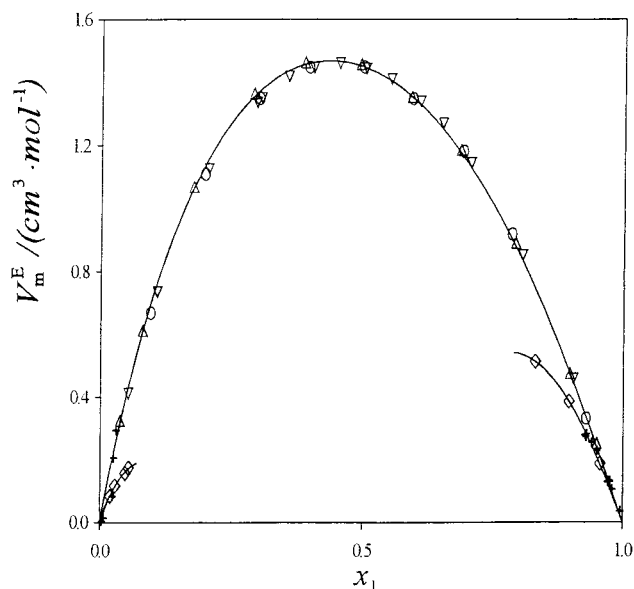


Figure 1. Curves of excess molar volumes V_m^E from Redlich–Kister eq 4 at 298.15 K for methanol (2) + cyclohexane (3) (\diamond , this work; +, Arce et al.⁴) and dimethyl carbonate (1) + cyclohexane (3) (\triangle , this work; ∇ , García de la Fuente et al.;⁵ \circ , Negadi et al.⁶ and fitted curves).

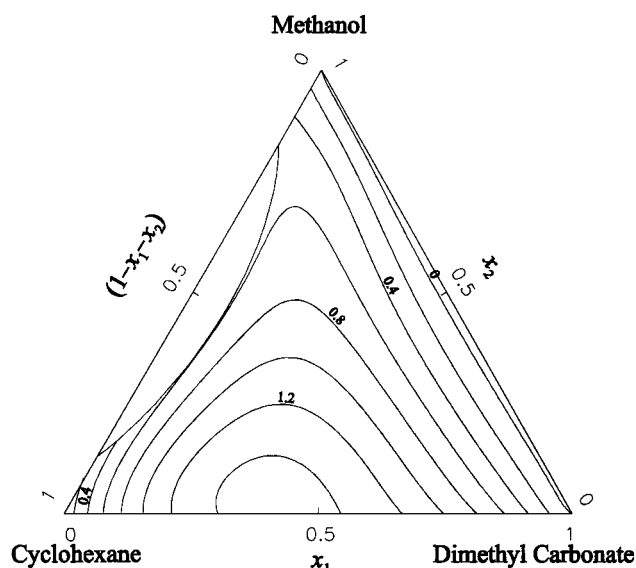


Figure 2. Curves of constant excess molar volumes V_m^E from Cibulka eq 5 at 298.15 K for dimethyl carbonate (1) + methanol (2) + cyclohexane (3).

Table 5. Liquid–Liquid Equilibrium of the Ternary System at 298.15 K

x_1^I	x_2^I	x_1^{II}	x_2^{II}
0	0.120	0	0.827
0.005	0.128	0.014	0.803
0.017	0.157	0.034	0.760
0.030	0.182	0.051	0.721
0.036	0.192	0.058	0.698
0.046	0.215	0.066	0.659
0.056	0.249	0.074	0.623

The predictive methods⁷ for excess properties of Kohler, Jacob, and Fitzner; Colinet, Tsao, and Smith; Toop; and Scatchard are based on additive binary contributions that determine ternary excess properties. For the ternary mixture Table 8 gives the difference between experimental and estimated excess values as a root-mean-square differ-

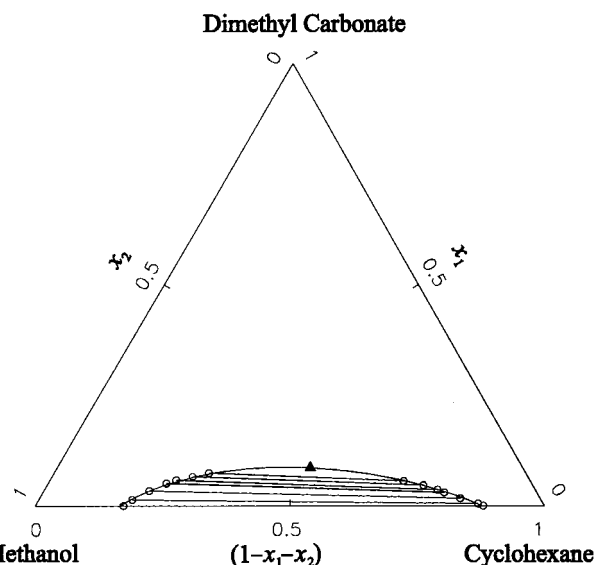


Figure 3. Experimental liquid–liquid equilibrium data of the ternary mixture dimethyl carbonate (1) + methanol (2) + cyclohexane (3) at 298.15 K: \circ – \circ , experimental tie lines and the binodal curve modeling by UNIQUAC; \triangle , plait point extrapolated.

Table 6. Comparison of Experimental Data with Literature of Mutual Solubility for Methanol (2) + Cyclohexane (3) at 298.15 K

x_2^I	x_2^{II}	refs
0.120	0.827	this work
0.128	0.825	Jones ¹³
0.1248	0.8280	Kiser ¹⁴
0.1250	0.8281	Yasuda et al. ¹⁵
0.1291	0.8284	Takeuchi et al. ¹⁶
0.1244	0.8285	Nagata and Katoh ¹⁷

Table 7. Correlating Parameters of the UNIQUAC Equation for the Ternary Mixture at 298.15 K and Root-Mean-Square Deviations σ

system	Δu_{ij}	Δu_{ji}
	$\text{kJ}\cdot\text{mol}^{-1}$	$\text{kJ}\cdot\text{mol}^{-1}$
dimethyl carbonate (1) + methanol (2)	–1364	–160
dimethyl carbonate (1) + cyclohexane (3)	–1338	152
methanol (2) + cyclohexane (3)	1272	21
$\sigma(x_1) = 0.007$	$\sigma(x_2) = 0.001$	

ence. The ternary excess properties of mixtures may be estimated from binary values applying

$$\Delta X_{ijk} = \sum_{i < j} (x_i x_j / x_i x_j) \Delta X_{ij}(x_i', x_j') \quad (7)$$

For each ternary mixture the mole fraction x' may be obtained from a triangular diagram by projecting the point representing the ternary mixture onto the corresponding binary axis, using different symmetric or asymmetric binary contributions to the ternary value. According to these rules, symmetry is understood as the contributions of the three binaries to the ternary excess, all contributing equally. Asymmetry is understood to indicate the different individual contributions of one of the binaries, attributed to the presence in the mixture of polar components. In general for dimethyl carbonate + methanol + cyclohexane the best estimations are given when the type b Tao–Smith asymmetric equation is applied for every derived property.

In Table 9, values of limiting pure partial excess molar volumes at 298.15 K for the binary mixtures are shown. These partial molar quantities are important in the study of the dependence of an extensive property on phase

Table 8. Root-Mean-Square Differences of the Experimental Results from the Predictions from Several Empirical Equations

eq	$\sigma(V_m^E)$ cm ³ ·mol ⁻¹	$\sigma(\Delta n_D)$	$\frac{\sigma(\Delta \kappa_S)}{T \cdot \text{Pa}^{-1}}$
Kohler	0.215	0.0017	12
Jacob-Fitzner	0.246	0.0016	13
Colinet	0.203	0.0016	11
Tsao-Smith ^a	0.184	0.0045	15
Tsao-Smith ^b	0.075	0.0003	5
Tsao-Smith ^c	0.228	0.0034	21
Scatchard ^a	0.199	0.0018	15
Scatchard ^b	0.258	0.0009	16
Scatchard ^c	0.220	0.0022	9
Toop ^a	0.182	0.0018	13
Toop ^b	0.247	0.0008	16
Toop ^c	0.220	0.0023	9

^a Cyclohexane 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.

Table 9. 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) + cyclohexane (3)	9.101	4.980
methanol (2) + cyclohexane (3)	5.002	4.549

composition at constant pressure and temperature, showing its trend with the composition. The partial excess molar volumes of a component in a multicomponent system can be obtained from the correlated parameters of excess volumes applying the Redlich-Kister and Cibulka equations. The limiting pure partial excess molar volumes should be determined by considering infinite dilution in the equation

$$\bar{V}_i^E = V_i^E + (\Delta V_i^E / \Delta x_i)_{P,T,x_j,j \neq i} - \sum_{k=1}^N x_k (\Delta V_k^E / \Delta x_k)_{P,T,x_j,j \neq k} \quad (8)$$

The limiting values obtained by eq 8 depend only on the correlation parameters.

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Registry Nos. Supplied by the Author: Dimethyl carbonate, 616-38-6; methanol, 67-56-1; cyclohexane, 110-82-7.

Literature Cited

- Rodríguez, A.; Canosa, J.; Tojo, J. Physical Properties of the Ternary Mixture Dimethyl Carbonate + Methanol + Benzene and Its Corresponding Binaries at 298.15 K. *J. Chem. Eng. Data* **1999**, *44*, 1298–1303.
- Redlich, O.; Kister, A. T. Thermodynamics of Nonelectrolytic Solutions. Algebraic Representation of Thermodynamic Properties and the Classification of Solutions. *Ind. Eng. Chem.* **1948**, *40*, 345–348.
- Cibulka, I. Estimation of Excess Volume and Density of Ternary Liquid Mixtures of Nonelectrolytes from Binary Data. *Collect. Czech. Commun.* **1982**, *47*, 1414–1419.
- Arce, A.; Blanco, A.; Antorrena, G.; Quintela, M. D. Propiedades Físicas de Exceso de Mezclas Ternarias. *An. Quím.* **1980**, *76*, 405–413.
- García de la Fuente, I.; González, J. A.; Cobos, J. C.; Casanova, C. Excess Molar Volumes for Dimethyl Carbonate + Heptane, Decane, 2,2,4-Trimethylpentane, Cyclohexane, Benzene, Toluene, or Tetrachloromethane. *J. Chem. Eng. Data* **1992**, *37*, 535–537.
- Negadi, L.; Blondel, A.; Mokbel, I.; Ait-Kaci, A.; Jose, J. *Int. DATA Ser.; Sel. Data Mixtures, Ser. A* **1993**, *21*, 183.
- Iglesias, M.; Orge, B.; Canosa, J.; Rodríguez, A.; Domínguez, M.; Piñeiro, M. M.; Tojo, J. Thermodynamic Behaviour of Mixtures Containing Methyl Acetate, Methanol and 1-Butanol at 298.15 K: Application of the ERAS Model. *Fluid Phase Equilib.* **1998**, *147*, 285–300.
- Abrams, D. S.; Prausnitz, J. M. *AIChE J.* **1975**, *21*, 116–121.
- Bevington, P. *Data Reduction and Error Analysis for the Physical Sciences*; McGraw-Hill: New York, 1969.
- Pal, A.; Dass, G.; Kumar, A. Excess Molar Volumes, Viscosities, and Refractive Indices of Triethylene Glycol Dimethyl Ether with Dimethyl Carbonate, Diethyl Carbonate, and Propylene Carbonate at 298.15 K. *J. Chem. Eng. Data* **1998**, *43*, 738–741.
- Das, A.; Frenkel, M.; Gadalla, N. M.; Marsh, K.; Wilhoit, R. C. *TRC Thermodynamic Tables*; Thermodynamic Research Center, Texas A&M University: College Station, TX, 1994.
- Riddick, J. A.; Bunger, W. B.; Sakano, T. K. *Organic Solvents Techniques of Chemistry*, 4th ed.; Wiley: New York, 1986; Vol. II.
- Jones, D. C.; Amstell, S. The critical solution temperature of the systems Methyl alcohol + cyclohexane as a means of detecting and estimating water in Methyl alcohol. *J. Chem. Soc.* **1930**, 1316–1321.
- Kiser, R. W.; Jonson, G. D.; Shelter, M. D. Solubilities of various hydrocarbons in Methanol. *J. Chem. Eng. Data* **1961**, *6*, 338–342.
- Yasuda, M.; Kawade, H.; Katayama, T. *Kagaku Kogaku Ronbunshu* **1975**, *1*, 172–176.
- Takeuchi, S. T.; Nitta, T.; Katayama, T. Effect of selfassociation on liquid-liquid equilibrium. Correlation of mutual solubility for Methanol-saturated hydrocarbons systems. *J. Chem. Eng. Jpn.* **1975**, *8*, 248–256.
- Nagata, I.; Katoh, K. Ternary Liquid-Liquid Equilibria for Acetonitrile-Ethanol-Cyclohexane and Acetonitrile-2-Propanol-Cyclohexane. *Thermochim. Acta* **1980**, *39*, 45–62.

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