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Thermodynamic Properties of the Ternary Mixture Acetone + Methanol + Ethanol at 298.15 K

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Speeds of sound, densities, and refractive indices of the ternary mixture acetone + methanol + ethanol have been measured at 298.15 K and atmospheric pressure. The excess molar volumes, changes of refractive indices, and isentropic compressibilities were determined from the experimental data. Several methods for predicting multicomponent derived properties from binary data were tested. The Peng−Robinson and Soave−Redlich−Kwong equations of state were also applied, with three different mixing rules, to calculate binary interaction parameters and predict ternary excess volumes, satisfactory results being obtained for this mixture.

1. Introduction

This paper continues our study of various homogeneous and heterogeneous azeotropic mixtures (Iglesias et al., 1996a, 1997, 1998; Orge et al., 1997). The aim of this research is the measurement of physical properties and study of liquid equilibrium of multicomponent mixtures, as well as the application of several predictive models to obtain theoretical predictions. The objective of the application of these techniques is identification of separation agents for binary azeotropic or binary mixtures with close boiling points in modified rectification processes.

This paper reports the measured densities, refractive indices and speeds of sound as well as excess and derived properties of the ternary mixture acetone + methanol + ethanol at 298.15 K and atmospheric pressure. The experimental data were fitted by means of the Cibulka (1982) equation.

The measured values have been also used to test several empirical predictive methods (Piñeiro et al., 1998), used to estimate multicomponent properties from the corresponding magnitude of the binary mixtures involved.

The equations of state of Peng−Robinson (Peng and Robinson, 1976) and Soave−Redlich−Kwong (Soave, 1972) were applied with several combination rules to calculate binary interaction parameters between the chemicals present in the mixture. After that, these parameters were applied for estimating the ternary excess molar volumes, no other multicomponent parameter being necessary. The results obtained agreed very close with the experimentaly determined volumetric data.

2. Experimental Section

The chemicals used for the preparation of the mixtures were Lichrosolv Quality and supplied by Merck. Before use, they were degassed by ultrasound technique, dried over molecular sieves type 3A (Aldrich catalog no. 20858-2), and kept in inert argon (N-55, less than 3 ppm in water) atmosphere. The purity of the chemicals was checked using gas chromatography, obtaining purities better than 99.8 mass %. The values measured of density, refractive index, and speed of sound for the pure chemicals were compared with those found in open literature, and the results are listed in Table 1. The mixtures were prepared by weight using a Mettler AE-240 balance, with an accuracy of $\pm 10^{-4}$. Densities and speeds of sound of the mixtures were determined by means of an Anton Paar DSA-48 density and sound analyzer, with an accuracy of $\pm 5 \times 10^{-5}$ g·cm^{-3} for densities and ± 1 m × s^{-1} for speeds of sound. Refractive indices were measured using an ABBEMAT-HP Dr. Kernchen automatic refractometer, the accuracy being $\pm 2 \times 10^{-5}$. The refractometer was thermostatyzed with a PolyScience controller bath model 9010, whose temperature stability was $\pm 10^{-2}$ K. The mole fractions were determined with an accuracy of $\pm 10^{-4}$. The accuracy obtained when measuring excess molar volumes, changes of refractive indices, and isentropic compressibilities were $\pm 5 \times 10^{-3}$ cm^3·mol^{-1}, $\pm 10^{-4}$, and $\pm$ 1 TPa^{-1}, respectively. Further details of the experimental procedures used and the mode of operation in our laboratory have already been published (Iglesias et al., 1996b).

Table 1. Comparison of Measured Pure Component Properties Data with Literature Values at 298.15 K

	ρ/(g·cm^{-3})		n_D		u/(m·s^{-1})	
component	exptl	lit.	exptl	lit.[b]	exptl	lit.
acetone	0.7844	0.784 29[a]	1.355 80	1.355 96	1161	1160.6[c]
methanol	0.7866	0.786 64[b]	1.326 45	1.326 52	1102	1102.0[d]
ethanol	0.7850	0.785 09[b]	1.359 22	1.359 41	1142	1142.4[c]

[a] Hnedkovsky and Cibulka (1993). [b] *TRC Thermodynamic Tables* (1994). [c] Papaloannou Panayiotou (1991). [d] Arce et al. (1996).

3. Results and Discussion

The values corresponding to densities, refractive indices, speeds of sound, and isentropic compressibilities (calculated by means of the Laplace equation $\kappa_S = \rho^{-1} \cdot u^{-2}$), are given in Table 2. The excess molar volumes, changes of refractive indices on mixing, and changes of isentropic compressibili-

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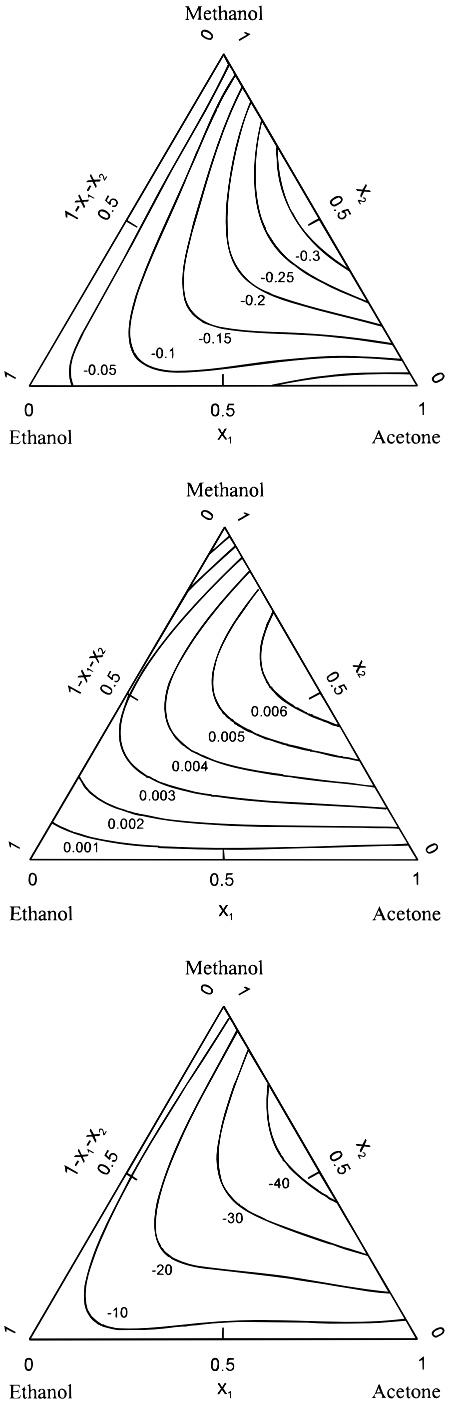

Figure 1. Curves of constant (a), excess molar volumes, (b) changes of refractive indices on mixing, and (c) changes of isentropic compressibilities for acetone (1) + methanol (2) + ethanol (3) at 298.15 K.

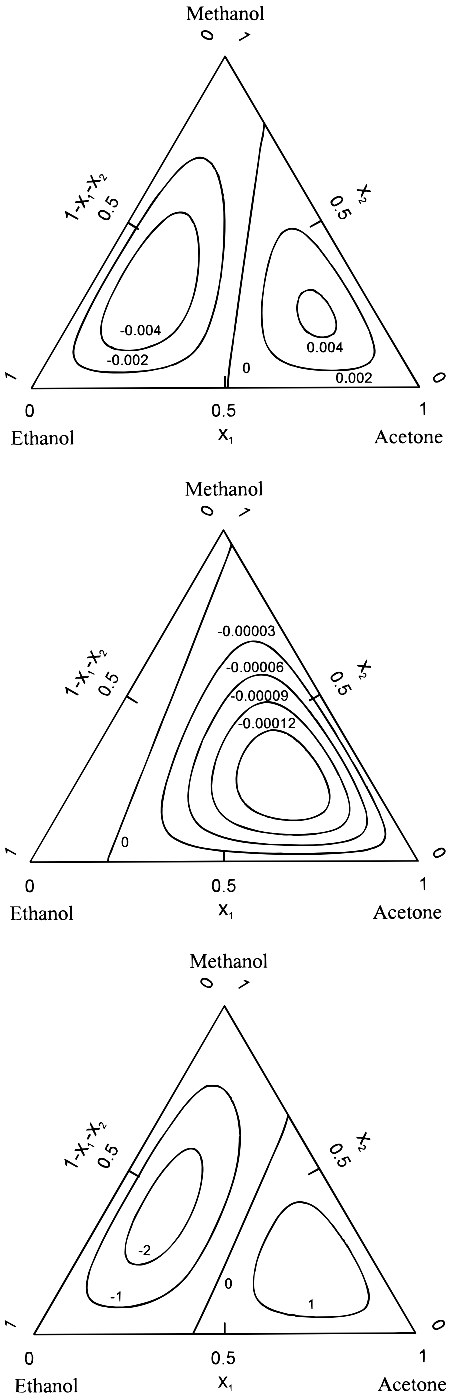

Figure 2. Curves of (a) $\delta V^{E}_{123} = V^{E} - V^{E}_{12} - V^{E}_{13} - V^{E}_{23}$, (b) $\delta(\delta n_{D,123}) = \delta n_{D} - \delta n_{D,12} - \delta n_{D,13} - \delta n_{D,23}$, and (c) $\delta(\delta\kappa_{S,123}) = \delta\kappa_{S} - \delta\kappa_{S,12} - \delta\kappa_{S,13} - \delta\kappa_{S,23}$ at 298.15 K for acetone (1) + methanol (2) + ethanol (3).

ties were calculated using the following expressions

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

$$\delta n_{D} = n_{D} - \sum_{i=1}^{N} x_i n_{Di} \tag{2}$$

$$\delta\kappa_{S} = \kappa_{S} - \sum_{i=1}^{N} x_i \kappa_{Si} \tag{3}$$

where ρ, n_D, and k_S stand for the density, refractive index, and isentropic compressibility of the mixture, and ρ_i, n_{Di}, and k_{Si} are the corresponding properties of pure components. N represents the number of components in the mixture. The excess and derived values are also presented in Table 2. These magnitudes were correlated using the Cibulka equation

$$\delta Q_{123} = \delta Q_{12} + \delta Q_{13} + \delta Q_{23} + x_1 \cdot x_2 \cdot x_3 \cdot (B_0 + B_1 \cdot x_1 + B_2 \cdot x_2) \tag{4}$$

where δQ_{12}, δQ_{13}, and δQ_{23} represent the binary contribu-

Table 2. Densities, Refractive Indices, Speeds of Sound, Isentropic Compressibilities, Excess Molar Volumes, Changes of Refractive Indices on Mixing, and Changes of Isentropic Compressibilities on Mixing, for Acetone (1) + Methanol (2) + Ethanol (3) at 298.15 K

x_1	x_2	ρ/(g·cm^{-3})	n_D	u/(m·s^{-1})	κ_S/(TPa^{-1})	V^E/(cm^3·mol^{-1})	δn_D	$\delta\kappa_S$/(TPa^{-1})
0.0312	0.0854	0.7856	1.357 24	1143	975	−0.039	0.0009	6
0.0319	0.2371	0.7859	1.353 59	1138	982	−0.045	0.0022	−10
0.0228	0.3871	0.7859	1.349 39	1133	992	−0.029	0.0029	−11
0.0241	0.5152	0.7861	1.345 22	1128	1000	−0.028	0.0030	−11
0.0230	0.6371	0.7864	1.341 41	1123	1008	−0.033	0.0031	−12
0.0220	0.7477	0.7865	1.337 45	1118	1018	−0.026	0.0028	−10
0.0230	0.8520	0.7868	1.333 41	1113	1026	−0.031	0.0022	−9
0.0211	0.9460	0.7870	1.329 30	1108	1035	−0.030	0.0012	−7
0.0906	0.0845	0.7858	1.357 30	1145	970	−0.058	0.0012	−9
0.1069	0.2373	0.7863	1.353 66	1142	975	−0.079	0.0026	−14
0.0984	0.3848	0.7867	1.349 81	1138	982	−0.089	0.0035	−18
0.0941	0.5162	0.7870	1.345 97	1133	990	−0.092	0.0040	−19
0.0898	0.6389	0.7874	1.341 94	1128	997	−0.100	0.0040	−21
0.0854	0.7523	0.7878	1.338 08	1124	1005	−0.108	0.0038	−21
0.0820	0.8514	0.7881	1.334 29	1119	1013	−0.110	0.0033	−21
0.1711	0.0854	0.7860	1.357 18	1148	965	−0.079	0.0013	−11
0.1644	0.2400	0.7867	1.353 67	1145	970	−0.112	0.0029	−17
0.1482	0.3971	0.7871	1.349 66	1140	978	−0.119	0.0040	−21
0.1464	0.5270	0.7876	1.345 90	1136	984	−0.135	0.0045	−25
0.1401	0.6533	0.7882	1.341 85	1131	991	−0.153	0.0045	−27
0.1323	0.7674	0.7886	1.338 01	1127	998	−0.158	0.0044	−27
0.1449	0.8115	0.7891	1.336 41	1126	999	−0.182	0.0043	−30
0.2132	0.3978	0.7875	1.349 62	1143	973	−0.153	0.0042	−25
0.2042	0.5315	0.7882	1.346 15	1139	978	−0.180	0.0050	−29
0.1942	0.6647	0.7889	1.342 10	1135	985	−0.202	0.0053	−32
0.1845	0.7785	0.7895	1.338 30	1131	991	−0.217	0.0052	−34
0.3119	0.0901	0.7862	1.356 90	1151	960	−0.105	0.0017	−13
0.2905	0.2523	0.7871	1.353 43	1148	964	−0.152	0.0035	−21
0.2792	0.4062	0.7880	1.349 88	1145	968	−0.194	0.0049	−28
0.2673	0.5429	0.7888	1.346 11	1142	972	−0.226	0.0056	−34
0.2552	0.6686	0.7895	1.342 42	1138	978	−0.248	0.0060	−38
0.3272	0.6397	0.7900	1.343 69	1143	969	−0.296	0.0066	−42
0.3386	0.5058	0.7889	1.347 33	1145	966	−0.248	0.0058	−35
0.3631	0.3501	0.7879	1.351 02	1149	962	−0.205	0.0045	−28
0.3916	0.0867	0.7862	1.356 71	1152	958	−0.111	0.0017	−13
0.3716	0.2552	0.7873	1.353 22	1150	960	−0.175	0.0036	−22
0.4710	0.0827	0.7861	1.356 55	1154	955	−0.110	0.0016	−12
0.4388	0.2663	0.7874	1.353 06	1152	957	−0.188	0.0041	−24
0.4245	0.4137	0.7886	1.349 67	1150	959	−0.253	0.0055	−33
0.4048	0.5592	0.7898	1.345 90	1147	962	−0.310	0.0064	−41
0.5489	0.0827	0.7860	1.356 37	1156	953	−0.108	0.0017	−12
0.5218	0.2612	0.7875	1.352 90	1154	953	−0.205	0.0040	−26
0.4881	0.4216	0.7889	1.349 67	1152	955	−0.282	0.0059	−36
0.6283	0.0983	0.7860	1.355 77	1157	950	−0.112	0.0019	−14
0.5919	0.2662	0.7876	1.352 70	1156	950	−0.219	0.0042	−27
0.5764	0.3701	0.7886	1.350 59	1155	950	−0.280	0.0055	−35
0.7089	0.0956	0.7859	1.355 70	1159	947	−0.110	0.0020	−14
0.6795	0.2756	0.7877	1.352 50	1159	945	−0.235	0.0046	−30
0.8035	0.0864	0.7856	1.355 45	1161	945	−0.091	0.0018	−13
0.1113	0.0548	0.7858	1.357 98	1147	968	−0.062	0.0009	−9
0.2422	0.0630	0.7860	1.357 58	1150	962	−0.086	0.0013	−11
0.0334	0.1480	0.7856	1.355 72	1141	978	−0.033	0.0015	−7
0.1337	0.1378	0.7861	1.356 01	1146	969	−0.078	0.0018	−13
0.2651	0.1642	0.7865	1.355 38	1149	963	−0.115	0.0024	−16
0.4071	0.1716	0.7867	1.354 97	1152	958	−0.142	0.0028	−18
0.4840	0.1805	0.7868	1.354 64	1154	955	−0.156	0.0030	−19
0.6456	0.1811	0.7868	1.354 26	1158	948	−0.171	0.0032	−21
0.0335	0.2335	0.7857	1.353 50	1138	982	−0.032	0.0020	−9
0.0337	0.3093	0.7858	1.351 52	1136	986	−0.031	0.0025	−10
0.1362	0.2948	0.7866	1.352 22	1142	975	−0.096	0.0031	−17
0.2612	0.3285	0.7874	1.351 55	1146	967	−0.161	0.0040	−24
0.4706	0.3532	0.7882	1.351 05	1152	956	−0.239	0.0050	−31

tion fitted with the Redlich−Kister expression for every binary mixture. The parameters of binary contributions were gathered in a previous work (Iglesias et al., 1997). B_i, $i = 0, 1, 2$, are the ternary fitting parameters that have been calculated applying the nonlinear algorithm due to Marquardt (Marquardt, 1963), and they are displayed in Table 3, as well as the root-mean-square deviations calculated according to the expression

$$\sigma = \left(\sum_i^{n_{DAT}} \frac{(z_{exp} - z_{cal})^2}{n_{DAT}} \right)^{1/2} \tag{5}$$

where z_{exp} is the experimental value, z_{cal} is the calculated value, and n_{DAT} is the number of experimental data points. Parts a, b, and c of Figure 1 show, respectively, the computed isolines corresponding to ternary derived properties. Figure 1a shows a contractive trend through the whole composition range, where a minimum value is reached in approximately equimolar binary composition of

Table 3. Parameters B_i of the Cibulka Equation and Root-Mean-Square Deviations σ

	Acetone (1) + Methanol (2) + Ethanol (3)			
V^E/(cm^3·mol^{-1})	$B_0 = -1.1325$	$B_1 = 1.6475$	$B_2 = 1.6184$	$\sigma = 0.0076$
δn_D	$B_0 = 0.0027$	$B_1 = -0.0137$	$B_2 = -0.0022$	$\sigma = 0.0001$
$\delta\kappa_S$/(TPa^{-1})	$B_0 = -167.5$	$B_1 = 398.1$	$B_2 = 46.9$	$\sigma = 0.4$

Table 4. Root-Mean-Square Deviations of the Experimental Results from the Predicted Results for Different Empirical Equations

	$\sigma(V^E/(\mathrm{cm^3{\cdot}mol^{-1}}))$	$\sigma(\delta n_D)$	$\sigma(\delta\kappa_S/(\mathrm{TPa^{-1}}))$
Kohler	0.005	0.000 1	1
Jacob−Fitzner	0.005	0.000 1	1
Colinet	0.005	0.000 1	1
Knobeloch	0.037	0.000 8	5
Tsao−Smith[a]	0.012	0.000 6	3
Tsao−Smith[b]	0.010	0.000 1	1
Tsao−Smith[c]	0.041	0.001 0	6
Scatchard[a]	0.011	0.000 2	2
Scatchard[b]	0.007	0.000 1	2
Scatchard[c]	0.009	0.000 1	1
Toop[a]	0.011	0.000 2	2
Toop[b]	0.007	0.000 1	1
Toop[c]	0.008	0.000 1	1
Mathieson−Tynne[a]	0.248	0.003 0	31
Mathieson−Tynne[b]	0.147	0.004 8	23
Mathieson−Tynne[c]	0.013	0.001 5	10
Hillert[a]	0.014	0.001 5	5
Hillert[b]	0.031	0.000 1	3
Hillert[c]	0.139	0.002 8	19

[a] Ethanol is the asymmetric component in the equation. [b] Methanol is the asymmetric component in the equation. [c] Acetone is the asymmetric component in the equation.

acetone + methanol owing to the hydrogen bonds between methanol and acetone molecules. In decreasing ethanol composition mixtures, lower values of excess molar volumes and an increasing trend toward pure methanol or acetone could be observed. The changes of refractive indices on mixing show positive values (Figure 1b), in agreement with this trend shown, a maximum appearing. The changes of isentropic compressibilities show a similar trend to excess molar volumes (Figure 1c).

The empirical predictive methods for derived properties of Kohler, Jacob and Fitzner, Colinet, Knobeloch, Tsao and Smith, Scatchard, Toop, Mathieson-Tynne, and Hillert (Piñeiro et al., 1998) calculate multicomponent properties by means of different additive binary contributions. Table 4 shows root-mean-square deviations computed using eq 5. The derived properties of multicomponent mixtures may be estimated using the corresponding binary data as follows

$$\delta Q_{ij} = \sum_{i<j} (x_i x_j / x'_i x'_j)\,\delta Q_{ij}(x'_i,\, x'_j) \qquad (6)$$

In this case, for each ternary mixture the mole fractions x' may be obtained in a triangular Gibbs diagram, projecting the considered ternary composition point on the binary axis. The possible combinations of symmetric and asymmetric geometric choice yield the expressions of the referred methods. Asymmetry can be regarded as the different individual contribution of one of the binary mixtures involved, caused by a polar or strong associative behavior in the multicomponent mixture. In Table 4 the deviations show that symmetric equations, except for Knobeloch, provide the best predictions for the set of magnitudes taken into consideration for this mixture. Low deviations are obtained by these methods owing to the scarce ternary contribution to derived magnitudes (Figure 2).

In what is referred to the equations of state, the facts of its high simplicity, low data requirements, and wide versatility in estimating different physical properties of both pure substances and multicomponent mixtures has attracted the general interest. The accuracy of the results obtained depend to a great extent on the combination of equations and mixing rules. A considerable number of equations of state are currently available, and most of them are adequate to obtain acceptable results, combined with simple rules, if some binary parameters are calculated from experimental data from the enclosed binary mixtures in the multicomponent system. In this case, the Soave−Redlich−Kwong (SRK) and the Peng−Robinson (PR), equations were selected and applied with combining rules where the a and b factors in the mixture are dependent on one or two fitting parameters. These equations can be expressed by the general equation

$$\mathrm{P} = \frac{RT}{V-b} - \frac{a}{(V+\delta_1 b)(V+\delta_2 b)} \qquad (7)$$

where δ_1 and δ_2 are parameters with the following values: $\delta_1 = 1$, $\delta_2 = 0$ for SRK and $\delta_1 = 1 + \sqrt{2}$, $\delta_2 = 1 - \sqrt{2}$ for PR equations. For a binary mixture at constant P and T, the excess molar volume can be expressed according to

$$V^E = \Delta V = V_m - \sum_{i=1}^{n} x_i V_i = \sum_{i=1}^{n} x_i(\bar{V}_i - V_i) \qquad (8)$$

where $\bar{V}_i$ is the partial molar volume defined by

$$\bar{V}_i = -\left(\frac{\partial P}{\partial n_i}\right)_{T,V,n}\left(\frac{\partial P}{\partial V_m}\right)^{-1}_{T,n} \qquad (9)$$

and can be calculated from the selected equation of state, attending to the i component and mixture molar volume dependence.

Three different combining rules were tested with these equations, showing different dependences of a and b

Table 5. Computed Binary Interaction Parameters by the Applied Mixing Rules on the Equations of State SRK and PR, and Root-Mean-Square Deviations of the Ternary Prediction Results

	Soave−Redlich−Kwong			Peng−Robinson		
mixture	R1	R2	R3	R1	R2	R3
acetone + methanol	4.938×10^{-2} (0.02)	1.753×10^{-2} -2.258×10^{-2} (0.009)	1.917 -6.373×10^{-3} -2.258×10^{-2} (0.01)	-5.764×10^{-2} (0.02)	2.610×10^{-2} -1.969×10^{-2} (0.009)	1.934 -6.351×10^{-3} -1.970×10^{-2} (0.009)
acetone + ethanol	1.758×10^{-2} (0.01)	3.224×10^{-2} -7.221×10^{-3} (0.006)	2.571×10^{-2} 2.187×10^{-5} -7.221×10^{-3} (0.007)	1.436×10^{-2} (0.01)	3.827×10^{-2} -5.578×10^{-3} (0.007)	2.327×10^{-2} 5.032×10^{-5} -5.578×10^{-3} (0.007)
methanol + ethanol	-1.013×10^{-2} (0.007)	5.317×10^{-2} 2.693×10^{-3} (0.004)	1.776×10^{-1} -4.174×10^{-4} 2.693×10^{-3} (0.004)	-1.065×10^{-2} (0.007)	6.145×10^{-2} 3.922×10^{-3} (0.004)	1.182×10^{-1} -1.904×10^{-4} 3.922×10^{-3} (0.004)
acetone + methanol + ethanol	(0.01)	(0.007)	(0.007)	(0.01)	(0.007)	(0.007)

parameters. A general equations could be expresed as

$$a = \sum_{i=1}^{n}\sum_{j=1}^{n} x_i x_j (1 - k_{ij} - l_{ij}T)(a_i a_j)^{1/2} \tag{10}$$

$$b = \sum_{i=1}^{n}\sum_{j=1}^{n} x_i x_j (1 - m_{ij})(b_i b_j)^{1/2} \tag{11}$$

where for mixing rule R1 $l_{ij} = m_{ij} = 0$, for mixing rule R2 $l_{ij} = 0$, and for mixing rule R3 k_{ij}, l_{ij}, $m_{ij} \neq 0$ these parameters being a constant value over the whole composition range for every binary mixture. The Marquardt algorithm was applied to calculate the parameters k_{ij}, l_{ij}, and m_{ij} in the cases mentioned above, using the experimentaly measured binary excess volumes, in combination with a Newton−Raphson method to solve the equation of state. Once these binary parameters were computed, the ternary excess values were predicted and compared with the experimental results. The parameters are given in Table 5, together with the root-mean-square deviations from the experimental data. The results displayed show that the predictions achieve a good accuracy, especially those calculated with mixing rules R2 and R3, and more precise than that usually offered by other theoretical methods when polar mixtures are involved. The equations of state reveal its usefulness as a predictive tool for excess volumes, as they have been validated in many other thermophysical magnitudes.

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