Mixing Properties for the Ternary Mixture Methyl *tert*-Butyl Ether + 1-Butanol + Decane at 298.15 K[†]

Manuel M. Piñeiro, Eva Mascato, Laura Mosteiro, and José Luis Legido*

Departamento de Física Aplicada, Facultade de Ciencias, Universidade de Vigo, Campus Lagoas-Marcosende, 36200 Vigo, Spain

Density and speed of sound at 298.15 K have been measured for the ternary mixture methyl *tert*-butyl ether (MTBE) + 1-butanol + decane and for the binary 1-butanol + decane. In addition, excess molar volumes and excess isentropic compressibilities were calculated from the speeds of sound, densities, cubic expansion coefficient, and literature heat capacities. Attending to the symmetry of experimental V^E and κ_S^E binary data, suitable fitting equations have been used. For the ternary data, different polynomial equations have been tested in order to minimize the set of parameters needed to correlate adequately. Experimental data were also used to test several empirical expressions for estimating ternary properties from experimental binary results.

Introduction

In the past few years, mixtures of ethers, alkanes, and alkanols have been the object of several investigations, as these mixtures are of technological importance, since the compounds involved, tertiary-alkyl ethers, either pure or mixed with alkanols, have been recommended as octane blending agents for gasolines. From a theoretical point of view, mixtures of alkanols and ethers are of interest due to their complexity, which is a consequence of the self-association of the alcohols, partially destroyed by the polar molecules of ether, and of the new intermolecular OH−O bonds created. A review paper of this kind of mixtures has been recently published,[1] where the lack of thermophysical properties of this kind of ternary mixtures is pointed out. A survey of the literature shows previously reported excess molar volume data for 1-butanol + decane,[2] but excess isentropic compressibility data and ternary data are not available.

This work reports experimental measurements of density and speed of sound for the binary mixture 1-butanol + decane and for the ternary mixture methyl *tert*-butyl ether (MTBE) + 1-butanol + decane at 298.15 K and atmospheric pressure. From experimental data, excess molar volumes and excess isentropic compressibilities were computed. According to Douhéret and Davis,[3] information about high energy chemical effects (chemical and intramolecular effects) can be obtained from excess molar volumes while excess isentropic compressibilities reflect lower energy effects (intermolecular and packing phenomena). Attending to the symmetry of the studied binary excess properties, suitable fitting equations have been used. Padè approximations were used to correlate excess molar volumes for 1-butanol + decane, whereas the remaining properties have been fitted with the Redlich−Kister equation.[4] For the ternary mixture, different polynomial equations[5−7] have been tested in order to minimize the set of parameters needed to correlate adequately the magnitude.

As the number of components in the mixture increases, the determination of thermodynamic properties becomes more laborious. Therefore, the applicability of predictive methods is of great interest for estimating ternary properties from the experimental data of the binaries involved. In this work several empirical equations[8−17] were evaluated to predict the ternary excess properties studied.

Experimental Section

MTBE (anhydrous) and 1-butanol (HPLC grade) were supplied by Aldrich, and decane (GC grade) was supplied by Sigma. The mole fraction purities stated by the manufacturers were better than 0.998 for MTBE and 1-butanol and better than 0.99 for decane. The substances were kept in argon (N-55) atmosphere, degassed, and stored over molecular sieves (Sigma, type 0.4 nm) to remove traces of water. The mole fraction of water in each of the liquids was determined by Karl Fischer titration to be <0.01 mol %. All compounds were checked by GLC analysis, and no significant impurities were found. Mixtures were prepared by mass using a Mettler AE-240 balance, with an accuracy of $\pm 5 \times 10^{-5}$ g, which leads to an estimated uncertainty in mole fraction of $\pm 5 \times 10^{-5}$. Precautions were taken during samples preparation, such as weighing liquids in increasing order of volatility and reducing to a minimum the vapor space in the vessels, to avoid losses by evaporation during manipulation and possible errors in mole fractions calculations.

Experimental values of density and speed of sound for the pure liquids at 298.15 K were compared with those found in the literature, being in a fairly good agreement, as shown in Table 1.

Density and speed of sound were measured at 298.15 K with an Anton Paar DSA-48 densimeter and sound analyzer, with an estimated uncertainty of $\pm 5 \times 10^{-5}$ g·cm^{-3} and $\pm 5 \times 10^{-1}$ m·s^{-1}, respectively. Both measuring cells are thermostated with the same solid-state thermostat, with a temperature uncertainty of $\pm 10^{-2}$ K. Apparatus calibration was performed periodically using air and Millipore quality water. Further information about the

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* Corresponding author. E-mail: xllegido@uvigo.es.

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Table 1. Comparison of Experimental Densities ρ, Speeds of Sound u, and Cubic Expansion Coefficients α with Literature Data for Pure Liquids at 298.15 K

	ρ/g·cm^{-3}		u/m·s^{-1}		α/10^{-3} K^{-1}		C_p/J·K^{-1}·mol^{-1}
compd	expt	lit.	expt	lit.	expt	lit.[j]	lit.[k]
MTBE	0.735 35	0.7353[a,b]	1036.1	1035[b] 1037[c]	1.430		187.49
1-butanol	0.805 67	0.8060[a] 0.8057[d]	1239.8	1239.3[e] 1239.39[f]	0.947	0.948	177.11
decane	0.726 09	0.72625[a] 0.72606[g]	1234.7	1234.37[h] 1234.34[i]	1.037	1.051	314.40

[a] Reference 18. [b] Reference 19. [c] Reference 20. [d] Reference 21. [e] Reference 22. [f] Reference 23. [g] Reference 24. [h] Reference 25. [i] Reference 26. [j] Reference 27. [k] Reference 29.

Table 2. Density ρ, Speed of Sound u, Excess Molar Volume V^E, and Excess Isentropic Compressibility κ_S^E at 298.15 K for 1-Butanol (1) + Decane (2)

x_1	ρ/g·cm^3	u/m·s^{-1}	V^E/cm^3·mol^{-1}	κ_S^E/TPa^{-1}
0.000 00	0.726 09	1234.7	0.000	0
0.048 48	0.727 38	1230.8	0.150	6
0.095 26	0.728 99	1228.9	0.216	9
0.204 46	0.733 33	1225.6	0.317	13
0.297 40	0.737 65	1223.9	0.365	16
0.394 87	0.742 88	1222.7	0.390	17
0.445 45	0.745 96	1222.5	0.386	17
0.492 84	0.749 07	1222.4	0.376	17
0.557 87	0.753 72	1222.7	0.362	17
0.593 72	0.756 50	1222.9	0.349	17
0.703 11	0.766 17	1224.6	0.291	14
0.790 14	0.775 35	1226.7	0.230	12
0.870 69	0.785 36	1230.4	0.159	8
0.947 96	0.796 76	1235.3	0.071	4
1.000 00	0.805 67	1239.8	0.000	0

Table 3. Density ρ, Speed of Sound u, Excess Molar Volume V^E, and Excess Isentropic Compressibility κ_S^E at 298.15 K for MTBE (1) + 1-Butanol (2) + Decane (3)

x_1	x_2	ρ/g·cm^3	u/m·s^{-1}	V^E/cm^3·mol^{-1}	κ_S^E/TPa^{-1}
0.075 67	0.051 36	0.727 86	1219.1	0.182	4
0.055 94	0.894 92	0.793 58	1224.2	−0.056	−13
0.127 03	0.096 86	0.729 94	1209.8	0.235	3
0.118 31	0.191 74	0.734 12	1208.4	0.264	4
0.116 09	0.293 65	0.739 37	1206.6	0.270	5
0.122 13	0.374 33	0.744 34	1204.6	0.242	2
0.110 75	0.487 33	0.751 92	1205.6	0.207	1
0.115 91	0.593 60	0.761 08	1205.2	0.114	−6
0.109 42	0.688 62	0.770 37	1207.9	0.032	−11
0.105 39	0.794 62	0.783 09	1211.9	−0.088	−21
0.170 56	0.092 62	0.730 12	1203.4	0.230	0
0.203 93	0.197 40	0.735 50	1195.1	0.227	−3
0.205 71	0.293 05	0.740 98	1192.9	0.192	−5
0.216 56	0.391 34	0.747 84	1189.5	0.117	−11
0.206 08	0.483 87	0.754 83	1190.4	0.047	−16
0.205 58	0.590 86	0.764 91	1190.9	−0.080	−26
0.206 90	0.693 01	0.776 86	1192.8	−0.243	−40
0.300 81	0.103 47	0.731 80	1182.1	0.225	−8
0.298 21	0.204 97	0.737 32	1179.5	0.171	−11
0.302 65	0.296 12	0.743 23	1176.6	0.091	−17
0.311 64	0.373 89	0.749 24	1173.5	−0.006	−24
0.311 60	0.483 92	0.759 00	1172.4	−0.164	−36
0.301 20	0.596 70	0.770 81	1174.9	−0.340	−52
0.396 85	0.109 00	0.733 13	1165.5	0.205	−14
0.403 00	0.196 76	0.738 60	1161.6	0.103	−20
0.397 16	0.303 24	0.746 11	1159.4	−0.034	−29
0.399 59	0.395 49	0.754 06	1157.4	−0.187	−41
0.404 77	0.494 66	0.764 63	1155.3	−0.408	−59
0.494 93	0.099 65	0.733 87	1147.5	0.174	−18
0.497 07	0.204 68	0.741 06	1143.2	0.002	−29
0.498 65	0.297 21	0.748 62	1141.6	−0.180	−44
0.497 94	0.402 83	0.758 95	1138.3	−0.416	−61
0.598 31	0.097 37	0.735 29	1126.3	0.116	−22
0.593 80	0.207 36	0.743 69	1123.7	−0.131	−39
0.596 69	0.301 18	0.752 52	1120.0	−0.378	−57
0.694 02	0.099 39	0.737 23	1106.0	0.025	−27
0.700 33	0.198 90	0.746 11	1100.8	−0.280	−47
0.795 98	0.102 58	0.739 89	1083.2	−0.118	−32
0.905 28	0.046 64	0.737 50	1058.4	−0.065	−18

experimental technique has been provided in a previous work.[7]

Results and Discussion

Excess properties, excess molar volumes V^E, and excess isentropic compressibilities κ_S^E were calculated using the following equation:

$$Q^E = Q - Q^{id} \tag{1}$$

where Q is either V/(cm^3·mol^{-1}) or $\kappa_S = \rho^{-1}u^{-2}$/(TPa^{-1}). The molar volume for an ideal mixture was calculated using the expression

$$V^{id} = \sum_{i=1}^{n} x_i V_i = \sum_{i=1}^{n} x_i M_i/\rho_i \tag{2}$$

where x_i, M_i, and ρ_i stand for the mole fraction, molar mass, and density of component i in the mixture, respectively, and n is the number of components in the mixture. The isentropic compressibility for an ideal mixture was evaluated using the expression suggested by Benson and Kiyohara[23] as follows:

$$\kappa_S^{id} = \sum_{i=1}^{n} \phi_i\{\kappa_{S,i} + TV_i\alpha_i^2/C_{p,i}\} - T\left(\sum_{i=1}^{n} x_i V_i\right)\left(\sum_{i=1}^{n} \phi_i\alpha_i\right)^2 \Big/ \sum_{i=1}^{n} x_i C_{p,i} \tag{3}$$

where ϕ_i is the volume fraction of component i in the mixture, stated in terms of the unmixed compounds, x_i is the corresponding mole fraction, T is the temperature, and $\kappa_{S,i}$, V_i, α_i, and $C_{p,i}$ are the isentropic compressibility, molar volume, cubic expansion coefficient, and molar heat capacity of component i, respectively. The values of α_i, calculated from a correlation function of density values at different temperatures[28] by analytical differentiation at 298.15 K, and the $C_{p,i}$ values, found in the literature,[29] are also reported in Table 1.

Densities ρ, speeds of sound u, V^E, and κ_S^E are listed in Table 2 for the binary mixture 1-butanol + decane and in Table 3 for the ternary mixture. Data for the binary mixtures MTBE + 1-butanol and MTBE + decane were published in previous works.[30,31]

Excess properties of the binary systems were correlated using the Padé approximant expression:

$$Q_{ij}^E = x_i x_j \frac{\sum_{p=1}^{m} A_p(2x_i - 1)^{p-1}}{1 + B_0(2x_i - 1)} \tag{4}$$

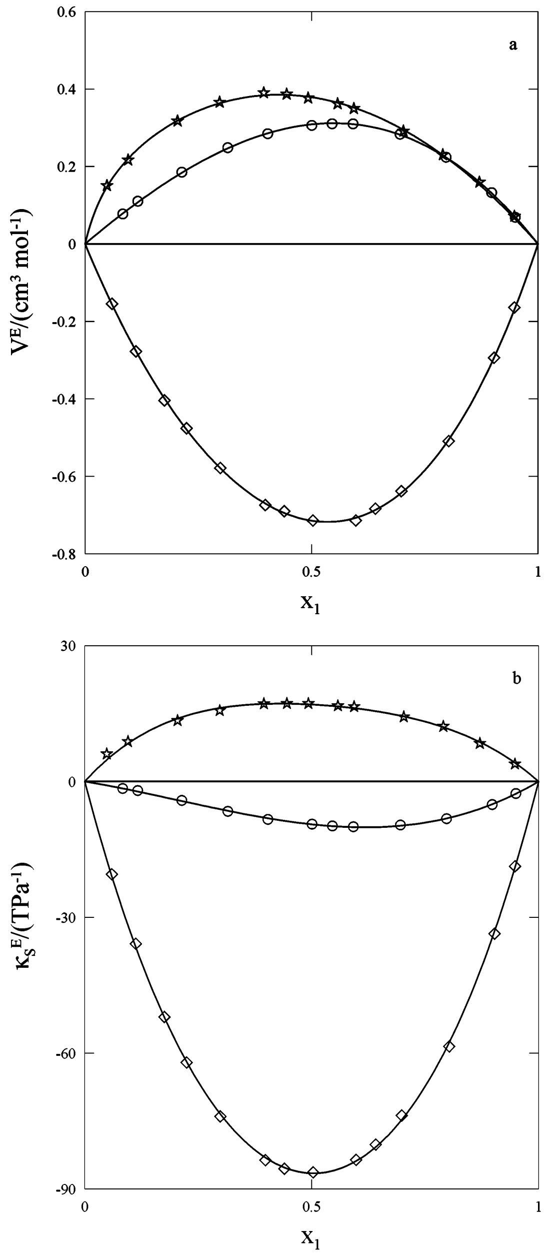

Figure 1. (a) V^E/cm^3·mol^{-1} and (b) κ_S^E/TPa^{-1} at 298.15 K for (◇) MTBE (1) + 1-butanol (2),[30] (○) MTBE (1) + decane,[31] (☆) 1-butanol + decane, and (—) eq 4.

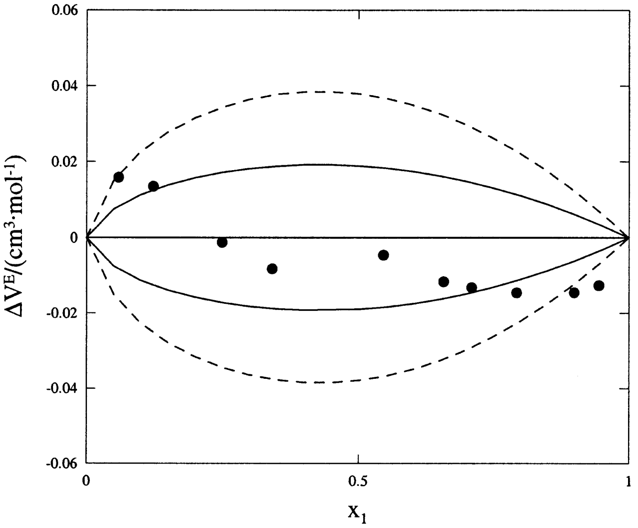

Figure 2. Deviations ΔV^E for 1-butanol (1) + decane (2) at 298.15 K (experimental values are shown through the zero value line; the plotted solid and dashed lines represent a 5% and a 10% deviation of V^E, respectively): ●, Kaur et al.[2]

Table 4. Parameters A_p and B_0 of Eq 4 and C_i of Eq 6, and Standard Deviations *s* for Excess Molar Volume and Excess Isentropic Compressibility

	B_0	A_1	A_2	A_3	*s*
	MTBE (1) + 1-Butanol (2)				
$V^{E\,a}$/cm^3·mol^{-1}		−2.858	−0.359	−0.319	0.004
$\kappa_S^{E\,a}$/TPa^{-1}		−346.1	−5.4	−41.5	0.6
	MTBE (1) + Decane (2)				
$V^{E\,b}$/cm^3·mol^{-1}		1.235	0.246		0.002
$\kappa_S^{E\,b}$/TPa^{-1}		−37.8	−21.3		0.1
	1-Butanol (1) + Decane (2)				
V^E/cm^3·mol^{-1}	0.899	1.515	1.011		0.004
κ_S^E/TPa^{-1}		68.0	−10.9	31.6	0.5

	C_0	C_1	C_2	*s*
	MTBE (1) + 1-Butanol (2) + Decane (3)			
V^E/cm^3·mol^{-1}	−1.622	3.298	3.903	0.005
κ_S^E/TPa^{-1}	118.9	150.6	438.6	0.5

[a] Reference 30. [b] Reference 31.

where x_i and x_j are the mole fractions of the components and A_p and B_i are the adjustable fitting parameters. An unweighted least-squares method was used to calculate these parameters. The number of parameters *m* was determined using the optimization F-test.[32] Figure 1 shows the experimental values of V^E and κ_S^E, plotted against molar fraction of the most volatile component of the binary mixture as well as the corresponding fitting curves. In Figure 2, the experimental values of excess molar volumes for the binary mixture 1-butanol + decane were compared with the literature data.

The ternary mixture derived properties were correlated using the equation

$$Q_{123}^E = Q_{12}^E + Q_{13}^E + Q_{23}^E + x_1x_2x_3\Delta_{123} \qquad (5)$$

where Q_{ij}^E is the binary contribution for each *ij* binary mixture, $x_3 = 1 - x_1 - x_2$, and $x_1x_2x_3\Delta_{123}$ is the ternary contribution which was correlated using the expression suggested by Cibulka[5]

$$\Delta_{123} = C_0 + C_1x_1 + C_2x_2 \qquad (6)$$

and the one proposed by Nagata−Tamura:[6]

$$\Delta_{123} = D_0 - D_1x_1 - D_2x_2 - D_3{x_1}^2 - D_4{x_2}^2 - D_5x_1x_2 - D_6{x_1}^3 - D_7{x_2}^3 - D_8{x_1}^2x_2 \qquad (7)$$

Ternary excess properties have been also correlated using the following expressions introduced in an earlier work:[7]

$$Q_{123}^E = E_1x_1x_2 + E_2x_2x_3 + E_3x_3x_1 + E_4{x_1}^2x_2 + E_5{x_2}^2x_3 + E_6{x_3}^2x_1 \qquad (8)$$

$$Q_{123}^E = F_1x_1x_2 + F_2x_2x_3 + F_3x_3x_1 + F_4{x_1}^2x_2 + F_5{x_2}^2x_3 + F_6{x_3}^2x_1 + F_7{x_1}^3x_2 + F_8{x_2}^3x_3 + F_9{x_3}^3x_1 \qquad (9)$$

The parameters A_p, B_0, and C_i of eqs 4 and 6 and the corresponding standard deviations are given in Table 4.

Table 5 presents a comparison among the standard deviations for ternary points only obtained with the dif-

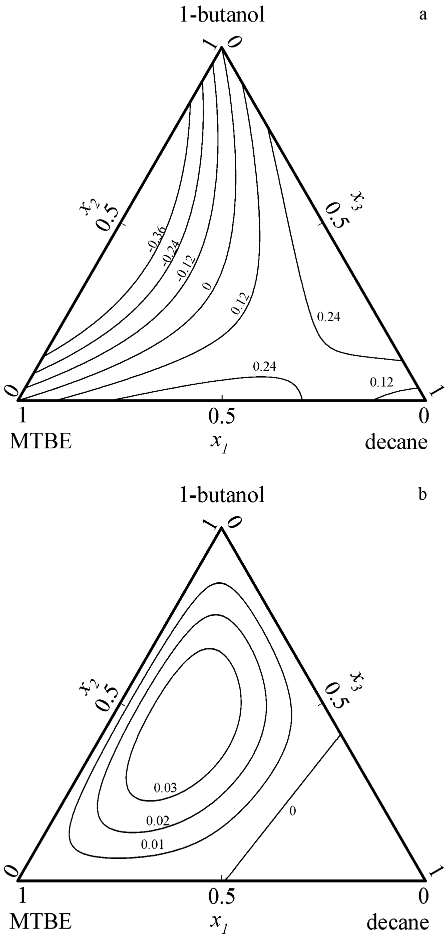

Figure 3. Isolines of (a) V^E/cm^3·mol^{-1} and (b) the ternary contribution in cm^3·mol^{-1} for MTBE (1) + 1-butanol (2) + decane (3): —, eq 6.

Table 5. Comparison of the Standard Deviations Obtained with the Correlation Equations Applied

	$s[V^E/(\text{cm}^3\cdot\text{mol}^{-1})]$	$s[\kappa_S^E/(\text{TPa}^{-1})]$
eq 6	0.005	0.5
eq 7	0.003	0.5
eq 8	0.009	1
eq 9	0.004	1

ferent correlation equations applied. Although eqs 8 and 9 do not include any ternary term, they yield low deviations when applied to ternary properties correlation using a reduced set of parameters.

The isolines of V^E and κ_S^E and their corresponding ternary contribution have been plotted in Figures 3 and 4, respectively.

Figures 3a and 4a show that both magnitudes present an isoline of ideal behavior. The negative region in the ternary diagrams is probably ascribable to the intermolecular OH−O bonds created between 1-butanol and MTBE molecules. For the mixture 1-butanol + decane, the predominant effect is the breakage of the self-association of the 1-butanol. The ternary contribution to the excess molar volume is practically negligible (Figure 3b) while for excess isentropic compressibility this contribution is relevant (Figure 4b).

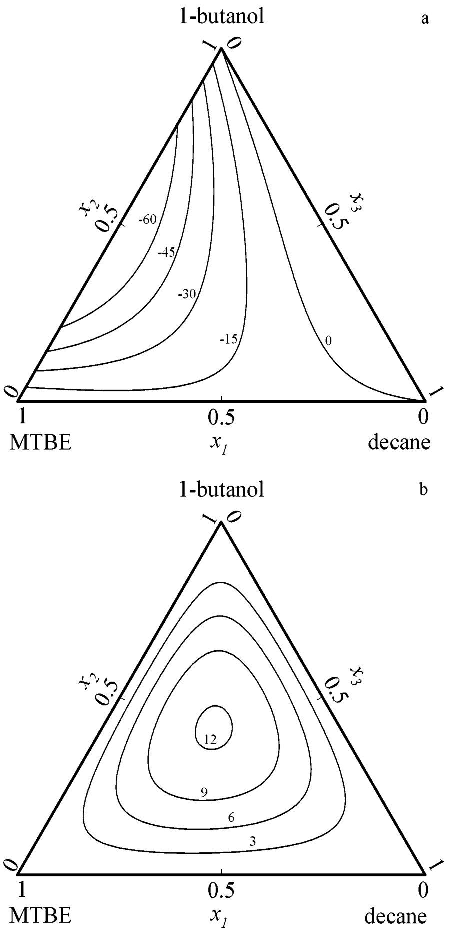

Figure 4. Isolines of (a) κ_S^E/TPa^{-1} and (b) the ternary contribution in TPa^{-1} for MTBE (1) + 1-butanol (2) + decane (3):—, eq 6.

Table 6. Root Mean Square Deviations of the Estimated Ternary Excess Molar Volume v_{123}^E and the Ternary Excess Isentropic Compressibility $\kappa_{S_{123}}^E$ from Experimental Data

	$\sigma[v_{123}^E/\text{cm}^3\cdot\text{mol}^{-1}]$			$\sigma[\kappa_{S_{123}}^E/\text{TPa}^{-1}]$		
Symmetric Equations						
Kohler	0.024			7.4		
Jacob−Fitzner	0.022			7.4		
Colinet	0.022			7.4		
Knobeloch−Schwartz	0.033			14.5		
Asymmetric Equations						
Chou	0.022			7.8		
Tsao−Smith	0.052^a	0.035^b	0.132^c	4.7^a	8.8^b	21.3^c
Toop	0.024^a	0.023^b	0.024^c	7.0^a	7.3^b	8.0^c
Scatchard	0.022^a	0.024^b	0.022^c	7.1^a	7.3^b	7.8^c
Mathieson−Thynne	0.022^a	0.023^b	0.023^c	7.2^a	7.3^b	7.7^c
Hillert	0.021^a	0.024^b	0.024^c	6.9^a	7.3^b	8.1^c

^{a} MTBE. ^{b} 1-Butanol. ^{c} Decane is respectively the asymmetric component in the equation.

Estimation Methods

Several empirical equations[8−17] have been applied to predict derived properties of ternary mixtures from the experimental data of the binaries involved. Using this kind of expressions, lower deviations are usually obtained for those excess properties where the ternary contribution is not important (Figures 3b). These equations are asymmetric when the numerical predictions depend on the arbitrary designation of component numbering, and they are symmetric otherwise. The equations of Kohler,[8] Jacob−Fitzner,[9] Colinet,[10] and Knobeloch−Schwartz[11] are symmetric, whereas those of Chou,[12] Tsao−Smith,[13] Toop,[14] Scatchard,[15] Mathieson−Thynne,[16] and Hillert[17] are asymmetric. The root mean square deviations between experimental and estimated values are shown in Table 6, where both symmetric and asymmetric equations perform the same for the excess molar volume, while the Tsao−Smith equation (with MTBE being the asymmetric component) performs the best for excess isentropic compressibility.

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