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Densities, Refractive Indices, and Derived Excess Properties of the System Methyl Acetate + Methanol + 2-Butanol at 298.15 K

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This work reports the experimental results of the densities and refractive indices of the ternary system methyl acetate + methanol + 2-butanol and the corresponding binary systems methyl acetate + 2-butanol and methanol + 2-butanol at 298.15 K and atmospheric pressure. A vibrating tube digital densimeter and a refractometer of synthetic sapphire prism (sodium D wavelengh) were used, the whole mole fraction range being covered. Excess molar volumes and changes of refractive index on mixing were determined from experimental data, and different estimation equations that are dependent of binary contributions were analyzed. Variable-degree polynomials have been fitted to the results. Values of densities and refractive indices are evaluated and compared by different methods. Excess partial molar volumes and those corresponding to the infinite dilution condition are also calculated.

1. Introduction

In continuation of our studies on the thermodynamic properties of binary and ternary mixtures methyl acetate + methanol + *n*-alkanols (Rodríguez *et al.*, 1996; Canosa *et al.*, 1997) and in view of the importance of the search for alternative entrainers in rectification processes for azeotropic mixtures, we report measurement of density and refractive index. The results were used to calculate excess molar volumes and changes of refractive index on mixing over the entire mole fraction range for methyl acetate + 2-butanol, methanol + 2-butanol, and the ternary system methyl acetate + methanol + 2-butanol at 298.15 K and atmospheric pressure.

Experimental values of density and refractive index of binary mixtures were correlated by Redlich−Kister (1948) equation. The binary contribution was used to correlate the experimental ternary results with the Cibulka (1982) equation. The experimental data of binary and ternary systems were used to test several different semiempirical relations. Ternary contributions were estimated and commented on in this work. The values obtained of excess molar volumes and ternary contributions were compared with the results for the mixture methyl acetate + methanol + 2-propanol (Canosa *et al.,* 1997).

2. Experimental Section

All the solvents used were from Merck with a LiChrosolv quality. 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 that fulfilled purchaser specifications. Their mass fraction purities were more than 0.998 for methyl acetate, methanol, and 2-butanol, with a maximum water contents of 6.8 $\times 10^{-3}$, 1.5×10^{-2}, and 8×10^{-3} %, respectively, determined using a Metrohm 737 KF coulometer. Densities and refractive indices at 298.15 K, which are in accordance with recommended and recent published values (Table 1), verified the purities.

A PolyScience controller bath model 9010 with a temperature stability of $\pm 10^{-2}$ K was used. Samples were prepared by mass using a Mettler AT-261 Delta Range balance with a precision of $\pm 10^{-5}$ g, covering the whole composition range of the mixture. The densities of mixtures and pure liquids were measured with an Anton Paar DMA-60/602 densimeter with a precision of $\pm 10^{-5}$ g·cm^{-3} and the refractive indices by the automatic refractometer ABBEMAT-HP Dr Kernchen with a precision of $\pm 10^{-5}$. Apparatus calibrations were realized periodically, testing fluids of known densities and refractive indices. The densimeter was calibrated with Millipore quality water and degassed and dried Fluka quality heptane. The accuracy in the calculation of excess molar volumes and changes of refractive indices was estimated as better than 10^{-3} for excess molar volumes and 2×10^{-5} for changes of refractive index on mixing. The accuracy in the mole fraction is 5 × 10^{-5}. The experimental technique and mode of operation has been described previously (Iglesias *et al.*, 1995).

3. Results and Discussion

Densities and refractive indices are given in Tables 2 and 3. The results were fitted to a polynomial equation. The density and refractive index for methyl acetate + methanol were reported earlier (Rodríguez *et al.*, 1996).

Excess molar volumes and changes of refractive index on mixing of binary and ternary mixtures were derived, respectively, from eqs 1 and 2.

$$V_{\mathrm{m}}^{\mathrm{E}} = \sum_{i=1}^{N} x_i M_i \,(\rho^{-1} - \rho_i^{\circ -1}) \qquad (1)$$

$$\Delta_{\mathrm{mix}} n_{\mathrm{D}} = n_{\mathrm{D}} - \sum_{i=1}^{N} x_i n_{\mathrm{D}i}^{\circ} \qquad (2)$$

In these equations, ρ is the density of the mixture, n_{D} is the refractive index of the mixture, and ρ_i° and $n_{\mathrm{D}i}^{\circ}$ are the density and refractive index of pure components.

Ternary results given in Table 3 were correlated with the Cibulka (1982) equation.

$$\Delta Q_{123} = \Delta Q_{12} + \Delta Q_{13} + \Delta Q_{23} + x_1 x_2 \,(1 - x_1 - x_2)(C_1 + C_2{\cdot}x_1 + C_3{\cdot}x_2) \qquad (3)$$

where ΔQ_{12}, ΔQ_{13}, and ΔQ_{23} are the parameters represent-

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

component	ρ(298.15 K)/(g·cm^{-3}) exptl	ρ(298.15 K)/(g·cm^{-3}) lit.	n_D(298.15 K) exptl	n_D(298.15 K) lit.
methyl acetate	0.926 74	0.927 3[a] 0.926 83[c]	1.358 50	1.358 90[a] 1.361 40[b]
methanol	0.786 65	0.786 64[a] 0.786 64[b]	1.326 45	1.326 52[a] 1.326 52[b]
2-butanol	0.802 37	0.802 6[a] 0.802 6[b]	1.394 88	1.395 0[a] 1.395 0[b]

[a] *TRC Thermodynamic Tables* (1994). [b] Riddick and Bunger (1986). [c] Lorenzi *et al.* (1995).

Table 2. Densities ρ, Refractive Indices n_D, Excess Molar Volumes V^E_m, and Changes of Refractive Index on Mixing δn_D for Binary Mixtures at 298.15 K

x_1	ρ/(g·cm^{-3})	n_D	V^E_m/(cm^3·mol^{-1})	$\Delta_{mix}n_D$
Methyl Acetate (1) + 2-Butanol (2)				
0.1075	0.811 22	1.390 0	0.324	−0.0010
0.2007	0.819 83	1.386 11	0.519	−0.0015
0.2978	0.829 64	1.382 41	0.654	−0.0016
0.3971	0.840 49	1.378 71	0.731	−0.0017
0.4911	0.851 54	1.375 22	0.753	−0.0018
0.5984	0.865 07	1.371 23	0.721	−0.0019
0.6996	0.878 86	1.367 58	0.631	−0.0018
0.8091	0.894 91	1.363 75	0.477	−0.0017
0.8915	0.908 01	1.361 17	0.304	−0.0013
Methanol (1) + 2-Butanol (2)				
0.1257	0.801 11	1.390 66	0.034	0.0044
0.2021	0.800 27	1.387 74	0.053	0.0067
0.2891	0.799 257	1.384 04	0.067	0.0089
0.3868	0.798 01	1.379 40	0.084	0.0110
0.5300	0.795 99	1.371 39	0.094	0.0128
0.6034	0.794 83	1.366 62	0.094	0.0130
0.6914	0.793 33	1.360 11	0.087	0.0125
0.8025	0.791 18	1.350 26	0.071	0.0103
0.8967	0.789 21	1.339 95	0.040	0.0064

ing the binary interactions in accordance with the Redlich−Kister expression:

$$\Delta Q_{ij} = x_i x_j \sum_{p=0}^{M} B_p (x_i - x_j)^p \qquad (4)$$

In this equation ΔQ_{ij} is the property, x is the mole fraction, B_p is a parameter, and M is the degree of the polynomic expansion. The unweighted least-squares method was used to fit the polynomials to the data. The degree of eq 4 was optimized by applying the F-test (Bevington, 1969). Curves of constant excess molar volumes and changes of refractive index on mixing for the ternary system have been plotted in parts a and b of Figure 1, respectively. The differences between the experimental values from literature and our results fitted for the methanol + 2-butanol binary system with eq 4 are showed in Figure 2.

Ternary systems exhibit positive values of the mixing properties over most of the composition diagram, except close to the binary systems methyl acetate + methanol where a change in sign occurs for excess volumes and for methyl acetate + 2-butanol for deviations of refractive indices. Positive or negative values of excess properties for that ternary mixture are influenced by the hydrogen bonds between high polar components and high molecular volume. In accordance with previous work (Canosa *et al.*, 1996), Figure 3 shows a comparison of excess molar volumes for the systems methyl acetate + methanol + 2-propanol and methyl acetate + methanol + 2-butanol when the mole fraction of methyl acetate is equal to methanol in the entire composition range for the third component.

Table 3. Densities, Refractive Indices, Excess Molar Volumes, and Changes of Refractive Index on Mixing for Methyl Acetate (1) + Methanol (2) + 2-Butanol (3) at 298.15 K

x_1	x_2	ρ/(g·cm^{-3})	n_D	V^E_m/(cm^3·mol^{-1})	$\Delta_{mix}n_D$
0.0492	0.2807	0.804 68	1.381 87	0.163	0.0080
0.0451	0.4290	0.803 15	1.374 84	0.148	0.0110
0.0416	0.7792	0.799 60	1.349 58	0.072	0.0095
0.0608	0.8802	0.802 85	1.337 55	0.023	0.0051
0.1686	0.3593	0.819 33	1.372 60	0.280	0.0084
0.1585	0.4775	0.819 00	1.366 38	0.215	0.0099
0.1490	0.5883	0.818 69	1.359 38	0.150	0.0102
0.1395	0.6774	0.818 03	1.352 87	0.099	0.0094
0.1284	0.7550	0.816 81	1.346 46	0.055	0.0079
0.1213	0.8202	0.816 14	1.340 10	0.015	0.0058
0.2645	0.3459	0.832 44	1.368 83	0.320	0.0072
0.2400	0.4728	0.831 64	1.362 62	0.218	0.0088
0.2231	0.5781	0.831 38	1.356 20	0.137	0.0090
0.2064	0.6686	0.830 65	1.349 76	0.073	0.0081
0.1859	0.7498	0.828 72	1.343 42	0.018	0.0066
0.3754	0.1942	0.843 65	1.371 70	0.475	0.0038
0.3414	0.3520	0.844 07	1.365 26	0.298	0.0069
0.3150	0.4735	0.844 17	1.359 07	0.181	0.0080
0.2859	0.5787	0.843 01	1.352 91	0.091	0.0080
0.2687	0.6659	0.843 15	1.346 56	0.019	0.0070
0.4746	0.1892	0.857 24	1.368 07	0.453	0.0034
0.4291	0.3442	0.857 60	1.361 94	0.257	0.0062
0.3912	0.4668	0.857 25	1.355 85	0.129	0.0071
0.3602	0.5676	0.856 87	1.349 91	0.029	0.0070
0.5620	0.1879	0.870 17	1.364 73	0.386	0.0031
0.5061	0.3412	0.870 36	1.358 76	0.182	0.0056
0.4623	0.4624	0.870 22	1.352 80	0.049	0.0064
0.6428	0.1911	0.883 16	1.361 50	0.282	0.0031
0.5821	0.3328	0.883 21	1.356 02	0.091	0.0051
0.7235	0.1850	0.896 30	1.358 72	0.157	0.0028
0.0526	0.1300	0.805 89	1.387 98	0.174	0.0039
0.2556	0.1264	0.827 17	1.379 42	0.495	0.0025
0.3243	0.1404	0.835 58	1.376 18	0.527	0.0027
0.4536	0.0790	0.849 88	1.373 55	0.626	0.0006
0.5484	0.0811	0.862 48	1.369 97	0.590	0.0006
0.6548	0.0764	0.877 27	1.366 22	0.510	0.0004
0.7383	0.0742	0.889 78	1.363 30	0.400	0.0004
0.3292	0.2567	0.839 27	1.370 70	0.405	0.0054
0.2020	0.2496	0.822 48	1.376 41	0.365	0.0060
0.6786	0.2354	0.892 73	1.357 76	0.123	0.0037
0.5122	0.2373	0.865 27	1.364 17	0.348	0.0042
0.1061	0.2039	0.811 12	1.382 68	0.270	0.0056
0.0965	0.3629	0.809 98	1.375 86	0.217	0.0093
0.0849	0.4879	0.808 28	1.369 44	0.174	0.0110
0.0814	0.5900	0.807 56	1.363 02	0.142	0.0115
0.0746	0.6825	0.806 23	1.356 40	0.108	0.0110
0.0653	0.8249	0.804 02	1.343 47	0.050	0.0074
0.8144	0.0931	0.903 71	1.359 88	0.210	0.0010

The parameters of the fitting equations (eqs 3 and 4 for excess values) and the corresponding root mean square deviations are given in Table 4.

Several methods to predict density and refractive indices of mixtures were applied to test their validity in high polar mixtures. The density of binary and ternary mixtures was estimated by the Hankinson−Brobst−Thompson (Hankinson and Thompson, 1979), Rackett (Rackett, 1970), Spencer and Danner modified Rackett (Spencer and Danner, 1972), Bradford−Thodos (Bradford and Thodos, 1968), Riedel (Riedel, 1954), Narsimham (Narsimham, 1967), and Yen−Woods (Yen and Woods, 1966) methods using expanded mixing rules proposed by those authors or the Kay (modified Prausnitz−Gunn combination) rule (Reid *et al.*, 1987). The refractive indices were compared with the predicted results for the mixing rules proposed by Lorentz−Lorenz, Dale−Gladstone, Arago−Biot (Tasic *et al.*, 1992), Eykman (Bottcher, 1952), Newton (Kurtz and Ward, 1936), Oster (Oster, 1948), and Eyring−John (Eyring and John, 1969) equations. The agreement between the experimental values and the calculated ones is illustrated by the figures given in Tables 5 and 6.

Table 4. Parameters of Equations 3 and 4 and Root Mean Square Deviations σ

		Methyl Acetate (1) + 2-Butanol (3)		
$V_m^E/(cm^3 \cdot mol^{-1})$	$B_0 = 3.0070$	$B_1 = -0.1389$	$B_2 = 0.4085$	$\sigma = 0.0013$
$\Delta_{mix}n_D$	$B_0 = -0.0072$	$B_1 = -0.0015$	$B_2 = -0.0074$	$\sigma = 0.00003$
		Methanol (2) + 2-Butanol (3)		
$V_m^E/(cm^3 \cdot mol^{-1})$	$B_0 = 0.3766$	$B_1 = 0.0888$		$\sigma = 0.0010$
$\Delta_{mix}n_D$	$B_0 = 0.0502$	$B_1 = 0.0192$	$B_2 = 0.0074$	$\sigma = 0.00004$
		Methyl Acetate (1) + Methanol (2) + 2-Butanol (3)		
$V_m^E/(cm^3 \cdot mol^{-1})$	$C_1 = -1.3631$	$C_2 = 0.9183$	$C_3 = 0.7708$	$\sigma = 0.0015$
$\Delta_{mix}n_D$	$C_1 = -0.0099$	$C_2 = 0.0032$	$C_3 = 0.0077$	$\sigma = 0.00005$

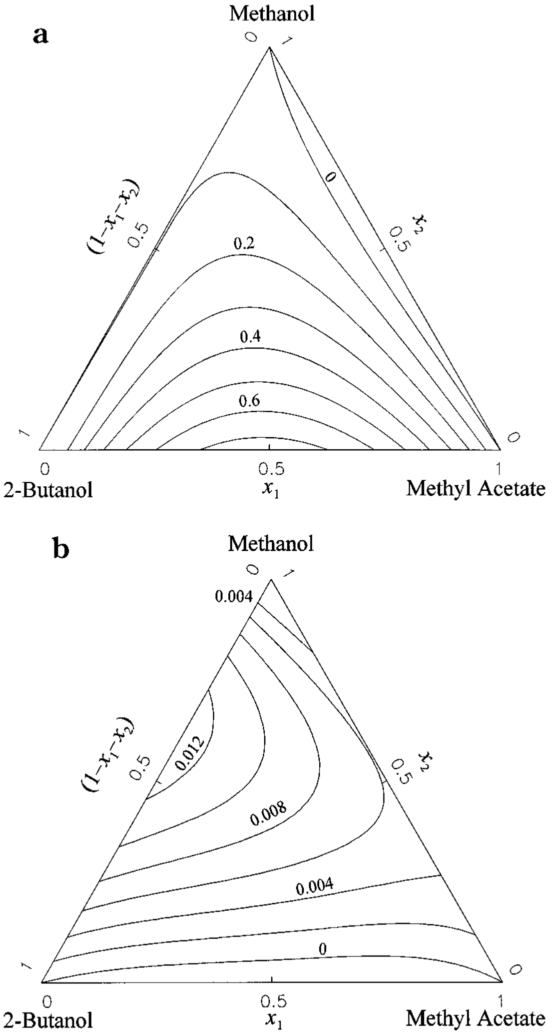

Figure 1. Curves (eq 3) of (a) excess molar volume and (b) changes of refractive index on mixing for the methyl acetate (1) + methanol (2) + 2-butanol (3) mixture at 298.15 K.

The excess molar volumes and changes of refractive index of mixing of more than two components may be estimated from binary results, applying eq 5:

$$\Delta Q_{ijk} = \sum_{i<j} (x_i x_j / x'_i x'_j) \Delta Q_{ij}(x'_i, x'_j) \qquad (5)$$

For each ternary mixture the mole fractions 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 criteria of binary contribution to the ternary value. In said rules symmetry is understood to be the contribution of the three binaries to the ternary excess, all three of which contribute equally. Asymmetry is understood to indicate the different individual contribution of one of the binaries, the latter being normally attributed to the polar component.

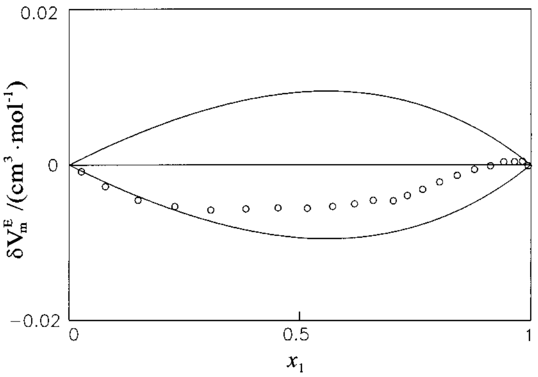

Figure 2. Deviations δV_m^E from eq 4 for methanol (1) + 2-butanol (2) at 298.15 K from available source: (○) Polak (1970); the zero line is our experimental values. The curves enclose deviations falling within ±0.1V^E.

Table 5. Root Mean Square Deviations (g·cm^{-3}) of the Experimental Results from Those Estimated for Densities (Hankinson–Brobst–Thompson (HBT), Rackett (R), Spencer and Danner Modified Rackett (SDR), Bradford–Thodos (BT), Riedel (Ri), Narsimham (N), and Yen–Woods (YW))

HBT	R	SDR	BT	Ri	N	YW
			Methyl Acetate (1) + 2-Butanol (3)			
0.010	0.022	0.064	0.004	0.035	0.006	0.009
			Methanol (2) + 2-Butanol (3)			
0.027	0.059	0.080	0.014	0.042	0.003	0.010
			Methyl Acetate (1) + Methanol (2) + 2-Butanol (3)			
0.020	0.054	0.060	0.013	0.032	0.004	0.012

The predictive methods for excess properties (Kohler, 1960; Jacob and Fitzner, 1977; Colinet, 1967; Tsao and Smith, 1953; Toop, 1965; Scatchard *et al.*, 1952) were used to determine ternary excess properties by means of additive binary contributions. In Tables 5, 6, and 7, the experimental and estimated properties and excess values are compared and corresponding root mean square deviations (σ) (eq 6) are shown. The value of the property and the number of experimental data are represented in eq 6 by z and n_{DAT}, respectively:

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

For density estimations in the ternary system, the Narsimham equation gives the smaller deviation, as well as in the methanol + 2-butanol mixture. The Bradford–Thodos equation gives the best agreement for methyl acetate + 2-butanol. For changes of refractive indices on mixing, the methanol + 2-butanol system shows the lowest deviations with the Eykman rule and the Dale–Gladstone

Table 6. Root Mean Square Deviations of the Experimental Results from the Prediction Results for Refractive Indices Lorentz–Lorenz (L–L), Dale–Gladstone (D–G), Arago–Biot (A–B), Eykman (Eyk), Newton (Nw), Oster (Os), and Eyring–Jhon (E–J) Equations

L–L	D–G	A–B	Eyk	Nw	Os	E–J
			Methyl Acetate (1) + 2-Butanol (3)			
0.002 53[a]	0.002 62[a]	0.002 62[a]	0.002 59[a]	0.002 71[a]	0.002 67[a]	0.002 57[a]
0.000 43[b]	0.000 24[b]	0.006 77[b]	0.000 28[b]	0.000 54[b]	0.000 31[b]	0.016 12[b]
			Methanol (2) + 2-Butanol (3)			
0.000 32[a]	0.000 61[a]	0.000 61[a]	0.000 51[a]	0.000 93[a]	0.000 77[a]	0.000 45[a]
0.000 18[b]	0.000 20[b]	0.000 99[b]	0.000 10[b]	0.000 56[b]	0.000 36[b]	0.002 74[b]
			Methyl Acetate (1) + Methanol (2) + 2-Butanol (3)			
0.001 15[a]	0.001 31[a]	0.001 31[a]	0.001 25[a]	0.001 50[a]	0.001 40[a]	0.001 22[a]
0.000 39[b]	0.000 16[b]	0.003 68[b]	0.000 22[b]	0.000 41[b]	0.000 22[b]	0.008 72[b]

[a,b] The molar volumes in the calculations are ($V^{\mathrm{id}} = \sum x_i V_i^{\circ}$)Δ (*a*) and ($V = (\sum x_i M_i^{\circ})/\rho$) (*b*).

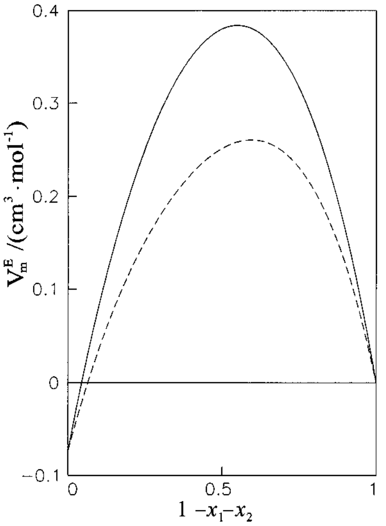

Figure 3. Curves (eq 3) of excess molar volume when $x_1 = x_2$ for mixtures: (—) methyl acetate (1) + methanol (2) + 2-butanol (3) and (- - -) methyl acetate (1) + methanol (2) + 2-propanol (3) at 298.15 K.

Table 7. Root Mean Square Deviations of the Experimental Results from the Prediction Results for Different Empirical Equations

	σ (V_m^E/(cm^3·mol^{-1}))	σ ($\Delta_{\mathrm{mix}} n_D$)
Kohler	0.015	0.0005
Jacob–Fitzner	0.012	0.0004
Colinet	0.015	0.0004
Tsao–Smith[a]	0.032	0.0024
Tsao–Smith[b]	0.126	0.0005
Tsao–Smith[c]	0.010	0.0016
Scatchard[a]	0.019	0.0005
Scatchard[b]	0.010	0.0001
Scatchard[c]	0.016	0.0008
Toop[a]	0.019	0.0006
Toop[b]	0.012	0.0002
Toop[c]	0.016	0.0008

[a–c] The asymmetric component in the equation is 2-butanol (*a*), methanol (*b*), and methyl acetate (*c*).

rule is best for methyl acetate + 2-butanol and methyl acetate + methanol + 2-butanol systems, all of them considering nonadditivity on mixing. In general for the ternary system the best estimates of excess volumes are given when symmetric equations and type b asymmetric equations are used except in the case of the type c Tsao–Smith equation. For deviations of refractive indices, symmetric equations are best; however, asymmetric equations give good predictions when a type b equation is used. Figure 4 shows the isolines corresponding to the ternary contribution, $V_{m,123}^E - V_{m,bin}^E$; in this one the interaction of the components when the composition is approximately equimolar is the highest. Figure 5 gives the ternary contribution for methyl acetate + methanol + 2-propanol and methyl acetate + methanol + 2-butanol systems.

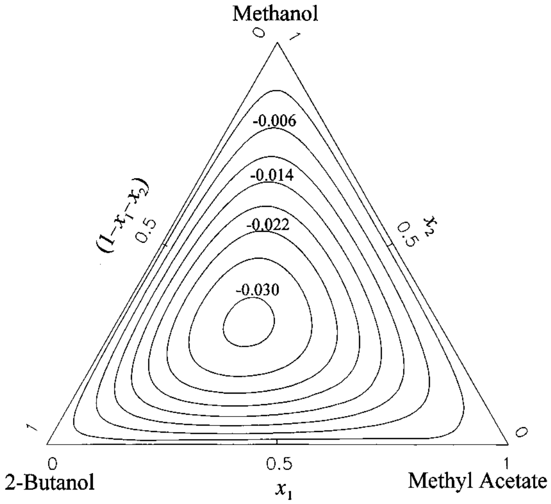

Figure 4. Curves (eq 3 minus eq 4) of constant ternary contribution to excess molar volumes of methyl acetate (1) + methanol (2) + 2-butanol (3) at 298.15 K.

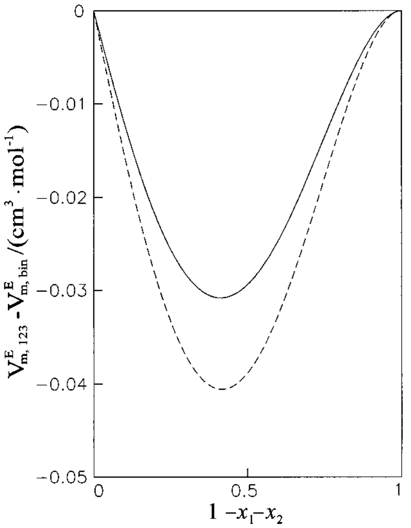

Figure 5. Curves (eq 3 minus eq 4) of constant ternary contribution to excess molar volumes when $x_1 = x_2$ for mixtures: (—) methyl acetate (1) + methanol (2) + 2-butanol (3) and (- -) methyl acetate (1) + methanol (2) + 2-propanol (3) at 298.15 K.

Figure 6a,b shows the volumes calculated for the Redlich–Kister equation as a function of mole fraction for the three binary mixtures. In general terms, it is observed as a positive trend corresponding to the binary mixture methyl acetate + 2-butanol and negative when the solvent is

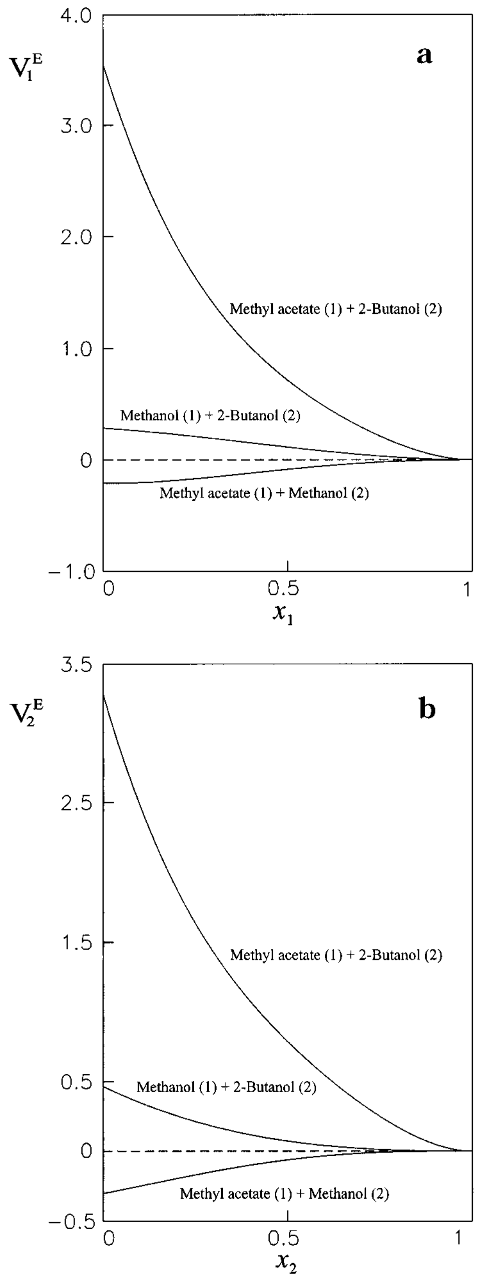

Figure 6. Partial excess molar volumes (eq 4) of methyl acetate (1) + 2-butanol (2), methyl acetate (1) + methanol (2), and methanol (1) + 2-butanol (2) mixtures at 298.15 K for (a) V_1^E and (b) V_2^E at 298.15 K.

Table 8. Partial Excess Molar Volumes at Infinite Dilution of the Binary Mixtures at 298.15 K

system	$V_1^{E,\infty}$/ (cm^3·mol^{-1})	$V_1^{E,\infty}$/ (cm^3·mol^{-1})
methyl acetate (1) + methanol (2)	−0.209	−0.301
methyl acetate (1) + 2-butanol (2)	3.554	3.277
methanol (1) + 2-butanol (2)	0.288	0.465

methanol. These results suggest the different packing of molecules when the chain of alcohols is increased. The binary mixture of the alcohols exhibits a small positive variation in the excess partial molar volume in the entire composition range. In Table 8, values of limiting partial excess molar volumes at 298.15 K for the binary mixtures are shown.

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Registry Nos. Supplied by the Author: Methyl acetate, 79-20-9; methanol, 67-56-1; 2-butanol, 78-92-2.

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