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Dynamic Viscosities of (Methyl Acetate or Methanol) with (Ethanol, 1-Propanol, 2-Propanol, 1-Butanol, and 2-Butanol) at 298.15 K

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Viscosities have been measured for the binary mixtures methyl acetate + (methanol, or ethanol, or 1-propanol, or 2-propanol, or 1-butanol and 2-butanol), and methanol + (ethanol, or 1-propanol, or 2-propanol, or 1-butanol and 2-butanol) and their viscosity deviations were evaluated at 298.15 K and atmospheric pressure over the entire composition range. Several semiempirical relations have been proposed to estimate dynamic viscosity, and the results were compared with the experimental data. The UNIFAC-VISCO group contribution method, Lobe's volume fraction exponential model, and the group contribution thermodynamics viscosity model (GC-UNIMOD) have been used to predict the dynamic viscosity.

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1. Introduction

The solution of many engineering problems concerning heat transfer, mass transfer, and fluid flow requires a knowledge of the viscosity of liquid mixtures and their dependence on composition and temperature. In this work, in the scope of a program to determine thermodynamic properties of new entrainers for homogeneous and heterogeneous extractive rectification, we present experimental values of dynamic viscosity for binary mixtures methyl acetate + (methanol, or ethanol, or 1-propanol, or 2-propanol, or 1-butanol and 2-butanol) and methanol + (ethanol, or 1-propanol, or 2-propanol, or 1-butanol and 2-butanol) at 298.15 K and atmospheric pressure.

Viscosity deviations are calculated from experimental densities and viscosities, covering the whole range of composition. We applied the correlation equations of Noda−Ishida (Noda and Ishida, 1977), Grunberg−Nissan (Nissan and Grunberg, 1949), Auslander (Auslander, 1964), Teja−Rice (Reid et al., 1987), UNIMOD (Cao et al., 1993a), and McAllister (McAllister, 1960), and a comparison between theoretical and experimental values is made. Dynamic viscosities for aforementioned binary mixtures have been predicted by the UNIFAC−VISCO (Gaston-Bonhomme et al., 1994) group contribution method and Lobe's volume fraction exponential model (Lobe, 1973), requiring only pure-component viscosities and the group contribution thermodynamics viscosity model (GC-UNIMOD) (Cao et al., 1993b).

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

component	η/(mPa·s) exptl	η/(mPa·s) lit.
methyl acetate	0.380	0.364[b]
		0.363[f]
methanol	0.553	0.5513[a]
		0.5450[b]
		0.5530[d]
		0.5486[f]
ethanol	1.105	1.0826[a]
		1.0825[f]
1-propanol	1.970	1.9430[a]
		1.9680[e]
		1.9653[f]
2-propanol	2.098	2.0436[a]
		2.0553[f]
		2.0820[g]
1-butanol	2.620	2.571[a]
		2.600[e]
		2.637[f]
2-butanol	3.115	2.998[a]
		3.084[c]
		3.174[f]

[a] *TRC Thermodynamic Tables* (1994). [b] Riddick and Bunger (1986). [c] Cea et al. (1994). [d] Wei-Rowley (1984). [e] Papaioannou and Panayiotu (1995). [f] DIPPR (1985). [g] Haase and Tillman (1995).

2. Experimental Section

The liquids used were from Merck with a LiChrosolv quality, except methyl acetate and 2-butanol, whose qualities are for synthesis and for analysis, respectively. The pure components were degassed ultrasonically, dried over freshly activated molecular sieves Type 3 Å and 4 Å (supplied by Aldrich) for several weeks before use, 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 99.8 mass % for methanol, ethanol, 1-propanol, 2-propanol and 1-butanol, more than 99 mass % for methyl acetate, and more than 99.5 mass % for 2-butanol, with a maxima water contents of 6.8×10^{-3}, 1.5×10^{-2}, 2.2×10^{-2}, 1.6×10^{-2}, 8.7×10^{-3}, 1.6×10^{-2}, and 8.15×10^{-3} mass %, respectively, determined using a Metrohm 737 KF coulometer. The purity of the solvents was assessed with recommended and published recent values (Table 1).

The mixtures were prepared by syringing weighed amounts of the pure liquids into stoppered bottles to prevent preferential evaporation, using a Mettler AT-261 Delta Range balance with a precision of 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 10^{-5} g·cm^{-3}, and calibrated with Millipore quality water and

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Table 2. Viscosities η and Viscosity Deviations $\Delta\eta$ for Binary Mixtures at 298.15 K

x_1	η/(mPa·s)	$\Delta\eta$/(mPa·s)	x_1	η/(mPa·s)	$\Delta\eta$/(mPa·s)
Methyl Acetate (1) + Methanol (2)					
0.0996	0.498	−0.038	0.6272	0.386	−0.058
0.2029	0.462	−0.056	0.7080	0.382	−0.049
0.3433	0.421	−0.073	0.7892	0.380	−0.036
0.4025	0.410	−0.073	0.8704	0.381	−0.021
0.4912	0.397	−0.072			
Methyl Acetate (1) + Ethanol (2)					
0.1146	0.833	−0.189	0.6077	0.438	−0.226
0.2045	0.695	−0.262	0.7045	0.418	−0.176
0.3207	0.574	−0.298	0.7989	0.399	−0.127
0.4245	0.506	−0.291	0.8802	0.390	−0.077
0.5004	0.467	−0.275			
Methyl Acetate (1) + 1-Propanol (2)					
0.0504	1.667	−0.223	0.6021	0.509	−0.504
0.1935	1.074	−0.588	0.6703	0.480	−0.424
0.3023	1.817	−0.672	0.7809	0.439	−0.289
0.4167	0.654	−0.653	0.8648	0.409	−0.186
0.5497	0.544	−0.552			
Methyl Acetate (1) + 2-Propanol (2)					
0.0598	1.642	−0.353	0.5621	0.504	−0.628
0.1684	1.101	−0.708	0.6773	0.448	−0.486
0.2671	0.830	−0.809	0.7619	0.417	−0.372
0.3634	0.671	−0.803	0.8660	0.394	−0.216
0.4646	0.571	−0.729			
Methyl Acetate (1) + 1-Butanol (2)					
0.0786	1.953	−0.491	0.5919	0.576	−0.718
0.1440	1.566	−0.731	0.6406	0.534	−0.651
0.2105	1.280	−0.868	0.7116	0.484	−0.542
0.2986	1.019	−0.932	0.7972	0.440	−0.394
0.4151	0.791	−0.899	0.9034	0.402	−0.194
0.4971	0.677	−0.829			
Methyl Acetate (1) + 2-Butanol (2)					
0.1056	1.854	−0.972	0.5697	0.559	−0.998
0.2017	1.275	−1.288	0.6653	0.485	−0.810
0.3076	0.921	−1.353	0.7801	0.429	−0.552
0.3884	0.771	−1.282	0.8760	0.411	−0.308
0.4935	0.640	−1.125			
Methanol (1) + Ethanol (2)					
0.0889	1.045	−0.011	0.5234	0.791	−0.025
0.1460	1.008	−0.016	0.6076	0.747	−0.023
0.2385	0.950	−0.023	0.7136	0.694	−0.017
0.3311	0.896	−0.026	0.8128	0.643	−0.013
0.4041	0.855	−0.027	0.9156	0.594	−0.006
Methanol (1) + 1-Propanol (2)					
0.1414	1.672	−0.098	0.5731	0.992	−0.166
0.2419	1.483	0.144	0.7216	0.825	−0.122
0.3419	1.315	−0.171	0.8038	0.744	−0.087
0.4462	1.159	−0.179	0.8805	0.662	−0.060
0.5137	1.067	−0.175			
Methanol (1) + 2-Propanol (2)					
0.0893	1.891	−0.069	0.6024	1.051	−0.116
0.2250	1.625	0.125	0.6852	0.945	−0.094
0.2835	1.518	−0.142	0.8272	0.772	−0.048
0.3984	1.332	−0.150	0.9058	0.675	−0.024
0.5178	1.160	−0.138			
Methanol (1) + 1-Butanol (2)					
0.1362	2.200	−0.138	0.6105	1.139	−0.219
0.2224	1.966	0.194	0.6930	0.999	−0.189
0.3020	1.768	−0.228	0.8012	0.833	−0.131
0.4090	1.528	−0.247	0.9004	0.690	−0.069
0.5154	1.312	−0.243			
Methanol (1) + 2-Butanol (2)					
0.1153	2.681	−0.139	0.5959	1.263	−0.325
0.2124	2.341	0.230	0.6710	1.102	−0.294
0.2972	2.063	−0.291	0.8045	0.852	−0.202
0.3974	1.765	−0.332	0.8927	0.711	−0.117
0.4714	1.563	−0.344			

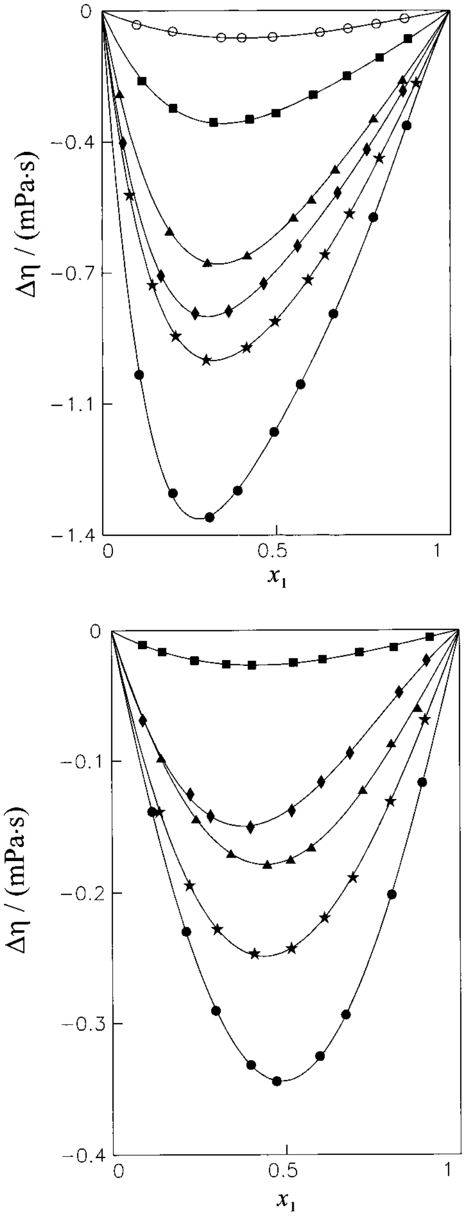

Figure 1. Curves (Redlich−Kister equation) of viscosity deviations for (a) ○ methyl acetate + methanol, ■ methyl acetate + ethanol, ▲ methyl acetate + 1-propanol, ♦ methyl acetate + 2-propanol, ★ methyl acetate + 1-butanol, ● methyl acetate + 2-butanol and (b) ■ methanol + ethanol, ▲ methanol + 1-propanol, ♦ methanol + 2-propanol, ★ methanol + 1-butanol, ● methanol + 2-butanol at 298.15K.

degassed and dried Fluka quality heptane as reference liquids. Viscosities were measured with an automated AMV 200 Anton Paar microviscosimeter and a precision better of 1%. The viscosimeter is based on the rolling-ball principle: a gold-covered steel ball rolls down inside an inclined, sample-filled glass capillary. The apparatus is equipped with an automatic timer (±0.01 s) and the time taken for the ball to roll a fixed distance between two magnetic sensors allows one to evaluate the viscosity of the mixture. The time measuring range is from 12 to 250 s, and the accuracy and the precision in this one are ±0.01 s and ±0.1 s, respectively. The capillary was placed in a block, thermostated with a PolyScience controller bath model 9010 with a temperature stability of $\pm10^{-2}$ K. Apparatus calibrations were realized periodically with Millipore quality water and degassed and dried Fluka quality octane and 1-pentanol. The accuracy in the viscosity was estimated as better than 2×10^{-2}. The accuracy in the mole fraction is 5×10^{-5}.

Table 3. Parameters (Eq 2) and Root-Mean-Square Deviations (Eq 3) σ for $\Delta\eta$/(mPa·s)

Methyl Acetate (1) + Methanol (2)					
$B_0 = -0.2834$	$B_1 = 0.1220$				$\sigma = 0.001$
Methyl Acetate (1) + Ethanol (2)					
$B_0 = -1.0854$	$B_1 = 0.7048$	$B_2 = -0.3319$			$\sigma = 0.002$
Methyl Acetate (1) + 1-Propanol (2)					
$B_0 = -2.3921$	$B_1 = 1.6549$	$B_2 = -0.8522$	$B_3 = 0.1533$		$\sigma = 0.002$
Methyl Acetate (1) + 2-Propanol (2)					
$B_0 = -2.7788$	$B_1 = 2.0260$	$B_2 = -1.5967$	$B_3 = 0.7363$		$\sigma = 0.001$
Methyl Acetate (1) + 1-Butanol (2)					
$B_0 = -3.3057$	$B_1 = 2.0678$	$B_2 = -1.5360$	$B_3 = 0.9324$	$B_4 = -0.1712$	$\sigma = 0.001$
Methyl Acetate (1) + 2-Butanol (2)					
$B_0 = -4.4647$	$B_1 = 3.2766$	$B_2 = -3.2220$	$B_3 = 2.3581$		$\sigma = 0.005$
Methanol (1) + Ethanol (2)					
$B_0 = -0.1043$	$B_1 = 0.0399$				$\sigma = 0.001$
Methanol (1) + 1-Propanol (2)					
$B_0 = -0.7041$	$B_1 = 0.1840$	$B_2 = 0.0510$			$\sigma = 0.002$
Methanol (1) + 2-Propanol (2)					
$B_0 = -0.5567$	$B_1 = 0.3289$				$\sigma = 0.002$
Methanol (1) + 1-Butanol (2)					
$B_0 = -0.9791$	$B_1 = 0.2578$				$\sigma = 0.001$
Methanol (1) + 2-Butanol (2)					
$B_0 = -1.3755$	$B_1 = 0.0819$	$B_2 = 0.1339$			$\sigma = 0.001$

3. Results and Discussion

The viscosity was measured at 298.15 K and atmospheric pressure, and the results are given in Table 2. In previous papers, we have measured the density of the binary liquid mixtures (Rodríguez et al., 1996, 1997; Canosa et al., 1997a,b). The viscosity deviations were calculated from dynamic viscosities and densities by eq 1

$$\Delta\eta = \eta - \sum_{i=1}^{N} (\eta_i^\circ x_i) \tag{1}$$

where η is the dynamic viscosity of the mixture and η_i° and x_i represent the viscosity of the pure component and mole fraction, respectively.

The results are given in Table 2 and are graphically presented in Figure 1a,b. The values were fitted to a Redlich−Kister (1948) expression

$$\Delta Q_{ij} = x_i x_j \sum_{p=0}^{M} B_p (x_i - x_j)^p \tag{2}$$

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 polynomial expansion. The parameters were computed by the least-squares method and optimized by applying the *F*-test (Bevington, 1969). They are shown in Table 3, together with the root-mean-square deviations (σ). The value of the property and the number of experimental data are represented in eq 3 by z and n_{DAT}, respectively:

$$\sigma = \left(\frac{\sum_{i}^{n_{\mathrm{DAT}}} (z_{\mathrm{exp}} - z_{\mathrm{pred}})^2}{n_{\mathrm{DAT}}} \right)^{1/2} \tag{3}$$

Parts a and b of Figure 1 show viscosity deviations for the binary mixtures methyl acetate + alcohols and methanol + alcohols, respectively, plotted against mole fraction together with the fitted curve. The viscosity deviations for the systems are negative over the entire composition range; this negative trend becomes broader when the alcohol chain length increases if the binary mixture is composed of an ester and an alcohol. However, when the mixture is methanol + ethanol, the viscosity deviations are almost zero.

Table 4. Root-Mean-Square Deviations (σ) for Comparison with Literature

systems	ref	σ ($\Delta\eta$/(mPa·s))
methyl acetate (1) + 1-propanol (2)	Acevedo et al.	0.026
methyl acetate (1) + 2-propanol (2)	Acevedo et al.	0.041
methanol (1) + ethanol (2)	Wei and Rowley	0.001
methanol (1) + 2-propanol (2)	Wei and Rowley	0.018
methanol (1) + 2-propanol (2)	Haase and Tillmann	0.028

Table 4 shows the root-mean-square deviations of the viscosity deviations for some recently published binary systems (Acevedo et al., 1990; Wei and Rowley, 1984; and Haase and Tillmann, 1995) and our viscosity deviations values.

The correlation equations of Auslander (Auslander, 1964), Grunberg−Nissan (Nissan and Grunberg, 1949), Noda−Ishida (Noda and Ishida, 1977), McAllister (McAllister, 1960), Teja−Rice (Reid et al., 1987), and UNIMOD (Cao et al., 1993a) were applied, the root-mean-square deviations being computed and gathered in Table 5.

On the basis of Eyring's theory of viscosities of pure liquids, many investigators have correlated the viscosities of binary liquid mixtures as follows:

$$\ln(\eta\cdot V) = \sum_{i=1}^{n} x_i \cdot \ln(\eta_i^\circ \cdot V_i^\circ) + \sum_{j=1}^{n} x_j \cdot \sum_{i=1}^{n} \frac{x_i \cdot W_{ij}}{\sum_{k=1}^{n} x_k \cdot \exp(-W_{ki}/R\cdot T)} \tag{4}$$

For the convenience of generalization to multicomponent mixtures, Noda and Ishida use the local composition for vapor−liquid equilibria, where W_{ij} is the adjustable parameter determined from the binary experimental data.

Table 5. Root-Mean-Square Deviations (σ) of the Experimental Results from Those Estimated for Viscosities (Auslander (AUSL), Grunberg−Nissan (G−N), Noda−Ishida (N−I), Mcallister (MCAL), Teja−Rice (T−R), and UNIMOD (UNIM))

AUSL	G−N	N−I	MCAL	T−R	UNIM
		Methyl Acetate (1) + Methanol (2)			
0.001	0.005	0.004	0.001	0.001	0.003
		Methyl Acetate (1) + Ethanol (2)			
0.002	0.016	0.016	0.002	0.013	0.003
		Methyl Acetate (1) + 1-Propanol (2)			
0.003	0.022	0.008	0.008	0.034	0.009
		Methyl Acetate (1) + 2-Propanol (2)			
0.004	0.034	0.006	0.006	0.062	0.007
		Methyl Acetate (1) + 1-Butanol (2)			
0.004	0.024	0.006	0.006	0.067	0.005
		Methyl Acetate (1) + 2-Butanol (2)			
0.008	0.057	0.010	0.012	0.133	0.010
		Methanol (1) + Ethanol (2)			
0.001	0.003	0.004	0.001	0.002	0.004
		Methanol (1) + 1-Propanol (2)			
0.003	0.008	0.011	0.004	0.005	0.018
		Methanol (1) + 2-Propanol (2)			
0.001	0.028	0.031	0.008	0.019	0.028
		Methanol (1) + 1-Butanol (2)			
0.001	0.021	0.029	0.006	0.004	0.028
		Methanol (1) + 2-Butanol (2)			
0.001	0.008	0.017	0.005	0.013	0.013

A statistical thermodynamic model (UNIMOD) for viscosity of pure liquids and liquid mixtures was developed by Cao et al. Local composition is introduced into the model. This one can be used to correlate viscosities of pure liquids and binary systems and to predict viscosities of multicomponent systems. For a liquid mixture, the dynamic viscosity equation is

$$\ln(\eta\cdot V) = \sum_{i=1}^{n}\phi_i\cdot\ln(\eta_i^{\circ}\cdot V_i^{\circ}) + 2\cdot\sum_{i=1}^{n}\phi_i\cdot\ln\frac{x_i}{\phi_i} - \sum_{i=1}^{n}\frac{q_i\cdot n_i\cdot\phi_i}{r_i}\cdot\sum_{j=1}^{n}\theta_{ji}\cdot\ln\tau_{ji} \qquad (5)$$

$$\theta_{ij} = \frac{\theta_j\cdot\tau_{ji}}{\sum_{l=1}^{n}\theta_l\cdot\tau_{li}} \qquad (6)$$

$$\tau_{ji} = \exp\left(-\frac{z}{2}\cdot\frac{U_{ji}-U_{ii}}{R\cdot T}\right) \qquad (7)$$

where V is the molar volume of the mixture, V_i° is the molar volume of component i, ϕ_i is the average segment fraction of component i, x_i is the mole fraction of component i, θ_{ji} is the local composition, θ_i is the average area fraction of component i, and τ_{ji} is the interaction parameter between sites j and i in the mixture. In the model, the interaction potential energy differences, $U_{ji} - U_{ii}$, for a liquid mixture represent the adjustable parameters. The viscosity data of binary system are used to determine them, and they can also determined from thermodynamic properties.

In general terms, low standard deviation values were obtained in almost the whole correlation. The system methyl acetate + 2-butanol shows the highest value for all equations owing to the high variation of dynamic viscosity with the composition. It is considered the best correlation equation for these binary mixtures Auslander's equation.

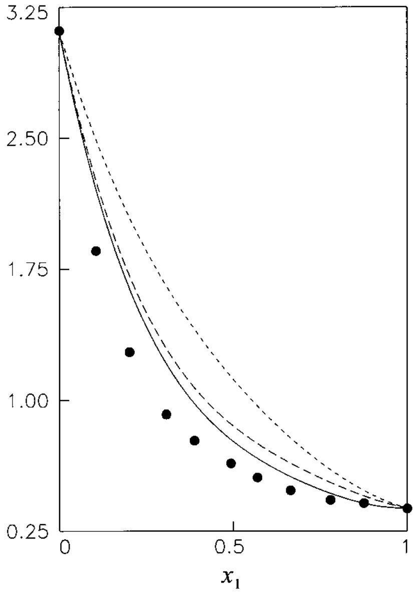

Figure 2. Comparison of binary methyl acetate + 2-butanol among experimental values ● and predictive ones (a) (—) UNIFAC-VISCO, (b) (— — —) Lobe and (c) (- - -) GC-UNIMOD at 298.15 K.

Table 6. Root-Mean-Square Deviations (σ) of Prediction Results of Viscosity for Binary Systems

systems	UNIFAC-VISCO	GC-UNIMOD	LOBE
methyl acetate (1) + methanol (2)	0.024	0.007	0.040
methyl acetate (1) + ethanol (2)	0.018	0.131	0.066
methyl acetate (1) + 1-propanol (2)	0.042	0.277	0.092
methyl acetate (1) + 2-propanol (2)	0.113	0.368	0.154
methyl acetate (1) + 1-butanol (2)	0.065	0.422	0.142
methyl acetate (1) + 2-butanol (2)	0.216	0.525	0.266
methanol (1) + ethanol (2)	0.029	0.018	0.030
methanol (1) + 1-propanol (2)	0.059	0.131	0.055
methanol (1) + 2-propanol (2)	0.031	0.090	0.020
methanol (1) + 1-butanol (2)	0.021	0.155	0.062
methanol (1) + 2-butanol (2)	0.013	0.195	0.040

In this paper, Table 6 shows the comparison of the experimental and calculated values of the viscosity in terms of root-mean-square deviations (σ) for the binary systems, using the predictive equations of UNIFAC-VISCO (Gaston-Bonhomme et al., 1994), Lobe (Lobe, 1973), and GC-UNIMOD (Cao et al., 1993b).

It is observed that in the predictive method of dynamic viscosity based on contribution groups UNIFAC-VISCO obtains the lowest deviation for the methanol + 2-butanol system; however, the binary systems methyl acetate with primary and secondary alcohols exhibit large root-mean-square deviations between experimental and calculated values. The same occurs when Lobe model is applied. However, the GC-UNIMOD predictive model for viscosities of binary mixtures shows the worst root-mean-square deviations, because the group interaction energy parameters used have been obtained from VLE data while the UNIFAC-VISCO model applies its own parameters. Figure 2 shows the comparison among the three predictive models and the experimental values for the binary mixture methyl acetate + 2-butanol, when the worst root-mean-square deviation is obtained. In general terms, the UNIFAC-VISCO model obtains the lowest root-mean-square deviations for the mentioned binary systems.

We must to take into account when we observe these results the fact that the UNIFAC-VISCO model, Lobe's volume fraction exponential, and the GC-UNIMOD model are predictive ones and they do not require the determination of adjustable parameters that would reduce this deviation.

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Registry Nos. Supplied by the Author: Methyl acetate, 79-20-9; methanol, 67-56-1; 1-propanol, 71-23-8, 2-propanol, 67-63-0, 1-butanol, 71-36-3, 2-butanol, 78-92-2.

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