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Density, Refractive Index, and Speed of Sound at 298.15 K, and Vapor−Liquid Equilibria at 101.3 kPa for Binary Mixtures of Methanol + Ethyl Butyrate and Vinyl Acetate + Ethyl Butyrate

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Densities, refractive indices, and speeds of sounds at 298.15 K, and isobaric vapor−liquid equilibria data at 101.3 kPa were reported for the binary mixtures containing methanol + ethyl butyrate and vinyl acetate + ethyl butyrate. Excess molar volumes, refractive index deviations, and changes of speed sound on mixing were calculated from the measurement results and fitted with Redlich−Kister polynomials. VLE experimental data were satisfactorily tested for thermodynamic consistency by means of a modified Dechema test. The activity coefficients were correlated with the Margules, van Laar, UNIQUAC, NRTL, and Wilson equations. The ASOG model was also used for prediction.

Introduction

Continuing our research on behavior and selection of possible entrainers to separate the methanol + vinyl acetate azeotropic mixture by extractive distillation, such as butanol,[1] 3-methyl-1-butanol,[2] butyl acetate and isobutyl acetate,[3] pentyl acetate and isopentyl acetate,[4] and propyl acetate and isopropyl acetate,[5] in this paper we will examine ethyl butyrate as the entrainer. We have measured vapor−liquid equilibria at 101.3 kPa for the methanol + ethyl butyrate and vinyl acetate + ethyl butyrate systems. Experimental measurements of the density, refractive index, speed sound, and vapor−liquid equilibrium data for binary mixtures of methanol + ethyl butyrate and vinyl acetate + ethyl butyrate were not found in the literature.

Experimental Section

Materials. Methanol (99.8 mol %) was supplied by Panreac and was used without further purification. Vinyl acetate (≥99 mol %) and ethyl butyrate (>98 mol %) from Fluka were purified by distillation in a laboratory column of 100 plates. The purities were checked by gas liquid chromatography and were higher than 99.7 mol %. All products were degassed using ultrasound and dried on molecular sieves (pore diameter 3 Å from Fluka). Densities, refractive indexes, speeds of sound, and normal boiling points of the pure substances are given in Table 1 and are compared with literature values of Riddick et al.[6]

Apparatus and Procedure. The still used to measure VLE data was a dynamic recirculating one described by Resa et al.[3] The equilibrium temperature was measured with a digital platinum 100 resistance thermometer with an accuracy of ±0.1 K. For the pressure measurement, a digital manometer regulator (Divatronic DT1 model, manufactured by Leybold) with an accuracy of ±0.1 kPa was used. Both vapor- and liquid-phase compositions for the two systems were determined by densimetry, refractometry, and speeds of sound. Densities were measured using an Anton Paar DMA 58 vibrating tube densimeter, at $T =$ 298.15 K with an accuracy of $\pm 0.00001\ \mathrm{g \cdot cm^{-3}}$, that had been calibrated at atmospheric pressure with twice distilled water and dry air. The temperature of the densimeter was maintained at 298.15 K with a precision of ±0.01 K by means of a semiconductor Peltier element and measured by a calibrated platinum resistance thermometer. Refractive indexes were measured with a Mettler RE50 refractometer, accuracy ±0.00001, whose temperature was controlled by a Peltier element, with a temperature precision of ±0.01 K. Speeds of sound were measured with an Anton Paar DSA 48 sound analyzer, accuracy ±0.1 m/s, whose temperature was also controlled with a Peltier element to within ±0.1 K. Prior to this, density-calibration, refractive index, and speed of sound curves for these systems were obtained to calculate the compositions of the vapor and liquid phases. The binary mixtures were prepared by directly weighing the constituent components with a SALTER electronic balance (model ER-182A) that has an accuracy of ±0.0001 g. Precautions were taken to mimimize evaporation losses during storage and preparation of the solutions. The estimated uncertainty in the determination of both liquid- and vapor-phase mole fractions is ±0.001.

Results and Discussion

Density, Refractive Index, and Speed of Sound. Table 2 lists the measured density ρ, refractive index n_D, and speed of sound u data at 298.15 K with the corresponding excess molar volume V^E, refractive index deviation δn_D, and speed of sound deviation δu for the binary mixtures of methanol + ethyl butyrate and vinyl acetate + ethyl butyrate.

The excess molar volumes of binary mixtures were calculated from density measurements applying the equation

$$V^E = x_1 M_1 (1/\rho - 1/\rho_1) + x_2 M_2 (1/\rho - 1/\rho_2) \qquad (1)$$

where ρ is the density of the mixture, ρ_1 and ρ_2 are the densities of the pure substances, M_1 and M_2 are the molar masses, and x_1 and x_2 are the molar fractions. Figure 1 illustrates the excess molar volumes of the two binary systems at 298.15 K.

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Table 1. Physical Properties of Pure Compounds: Densities ρ, Refractive Indexes n_D, and Speeds of Sound u at 298.15 K, and Normal Boiling Points T_b

	ρ/kg·m^{-3}		n_D		u/m·s^{-1}		T_b/K	
	obs	lit.[a]	obs	lit.[a]	obs	lit.[a]	obs	lit.[a]
methanol	786.56	786.37	1.326 34	1.326 2	1101.8	not available	337.9	337.696
vinyl acetate	925.59	not available	1.392 53	1.3934	1115.6	not available	346.0	345.7
ethyl butyrate	873.47	873.94	1.389 80	1.3928	1173.4	not available	394.5	394.70

[a] Riddick et al.[6]

Table 2. Densities, Refractive Indexes, and Speeds of Sound for Methanol (1) + Ethyl Butyrate (2) and Vinyl Acetate (1) + Ethyl Butyrate (2) at 298.15 K with Excess Molar Volume V^E, Refractive Index Deviation δn_D, and Speed of Sound Deviation δu

x_1	ρ / kg·m^{-3}	10^6V^E / m^3·mol^{-1}	n_D	δn_D	u / m·s^{-1}	δu / m·s^{-1}
Methanol (1) + Ethyl Butyrate (2)						
0.045	872.24	−0.0004	1.389 00	0.0020	1172.6	2.4
0.098	870.68	−0.0014	1.387 94	0.0044	1171.3	4.9
0.144	869.24	−0.0048	1.386 88	0.0062	1169.9	6.8
0.194	867.58	−0.0090	1.385 70	0.0082	1168.0	8.5
0.261	865.08	−0.0118	1.383 86	0.0106	1165.2	10.5
0.307	863.17	−0.0123	1.382 69	0.0124	1163.1	11.7
0.338	861.83	−0.0139	1.381 61	0.0133	1161.6	12.4
0.393	859.21	−0.0160	1.379 62	0.0148	1158.9	13.7
0.455	855.95	−0.0175	1.377 29	0.0163	1155.5	14.7
0.503	853.11	−0.0190	1.375 18	0.0173	1152.7	15.3
0.528	851.47	−0.0189	1.374 12	0.0178	1151.2	15.6
0.604	846.02	−0.0177	1.370 02	0.0185	1146.1	15.9
0.658	841.42	−0.0161	1.366 66	0.0186	1142.1	15.8
0.704	836.98	−0.0134	1.363 58	0.0185	1138.2	15.2
0.749	832.04	−0.0095	1.359 76	0.0175	1134.2	14.5
0.802	825.40	−0.0057	1.354 90	0.0160	1128.8	12.8
0.851	818.27	−0.0037	1.349 69	0.0139	1123.3	10.8
0.900	809.68	−0.0020	1.343 43	0.0108	1116.9	7.9
0.950	799.40	−0.0004	1.335 92	0.0064	1109.9	4.5
Vinyl Acetate (1) + Ethyl Butyrate (2)						
0.043	874.99	0.0077	1.389 90	−0.000 02	1171.1	0.3
0.083	876.49	0.0137	1.389 95	−0.000 08	1169.1	0.6
0.169	879.79	0.0261	1.390 10	−0.000 16	1164.6	1.1
0.191	880.66	0.0291	1.390 13	−0.000 19	1163.5	1.2
0.246	882.89	0.0354	1.390 21	−0.000 26	1160.6	1.5
0.307	885.50	0.0391	1.390 34	−0.000 30	1157.3	1.7
0.348	887.31	0.0420	1.390 43	−0.000 32	1155.1	1.9
0.409	890.12	0.0449	1.390 56	−0.000 36	1151.7	2.0
0.445	891.83	0.0460	1.390 63	−0.000 38	1149.7	2.1
0.497	894.41	0.0471	1.390 75	−0.000 41	1146.7	2.1
0.552	897.20	0.0483	1.390 87	−0.000 44	1143.5	2.0
0.594	899.46	0.0468	1.390 97	−0.000 45	1140.9	1.9
0.649	902.46	0.0465	1.391 13	−0.000 44	1137.6	1.7
0.693	904.99	0.0449	1.391 26	−0.000 43	1134.9	1.6
0.749	908.38	0.0393	1.391 46	−0.000 39	1131.5	1.4
0.807	912.02	0.0330	1.391 69	−0.000 31	1127.9	1.2
0.863	915.69	0.0270	1.391 91	−0.000 25	1124.4	0.9
0.899	918.14	0.0224	1.392 06	−0.000 19	1122.1	0.7
0.952	921.98	0.0121	1.392 31	−0.000 09	1118.6	0.2

The changes of refractive index δn_D at 298.15 K from the linear additive value of the mole fraction are obtained by

$$\delta n_D = n_D - (x_1 n_{D1} + x_2 n_{D2}) \qquad (2)$$

where n_D is the refractive index of the mixture and n_{D1} and n_{D2} are the refractive indices of the pure compounds. The plot of δn_D versus the mole fraction of the more volatile compound of each binary system x_1 is given in Figure 2.

In the same way, the changes of speeds of sound on mixing were calculated by the equation

$$\delta u = u - (x_1 u_1 + x_2 u_2) \qquad (3)$$

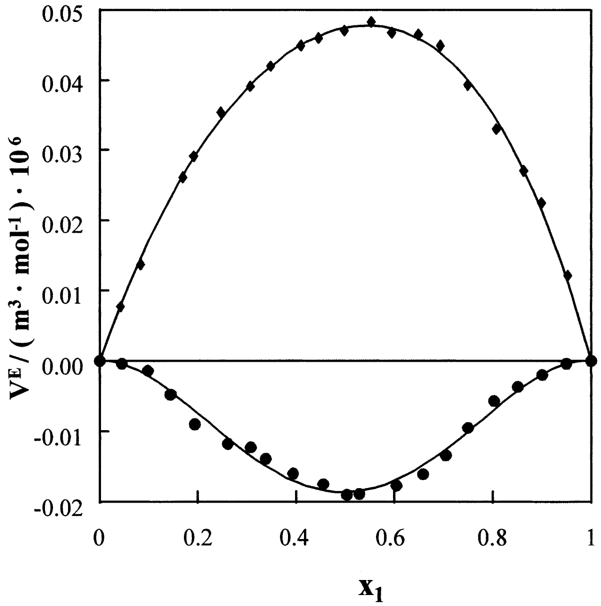

Figure 1. Excess molar volumes of mixtures of methanol (1) + ethyl butyrate (2) (●) and vinyl acetate (1) + ethyl butyrate (2) (♦) at 298.15 K.

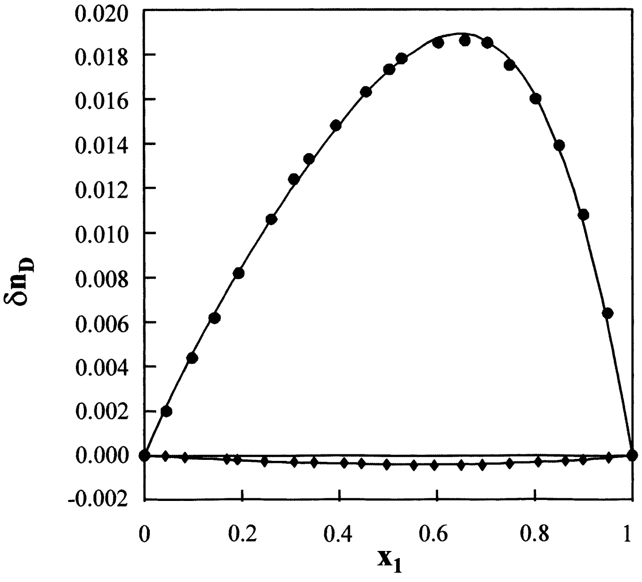

Figure 2. Change of refractive indices on mixing of methanol (1) + ethyl butyrate (2) (●) and vinyl acetate (1) + ethyl butyrate (2) (♦) at 298.15 K.

where u is the speed of sound of the mixture and u_1 and u_2 are the speeds of sound of the pure compounds. The plot of δu versus the mole fraction of the more volatile compound of each binary system x_1 is given in Figure 3.

Excess molar volumes and changes of refractive index and speeds of sound on mixing of the binary systems were

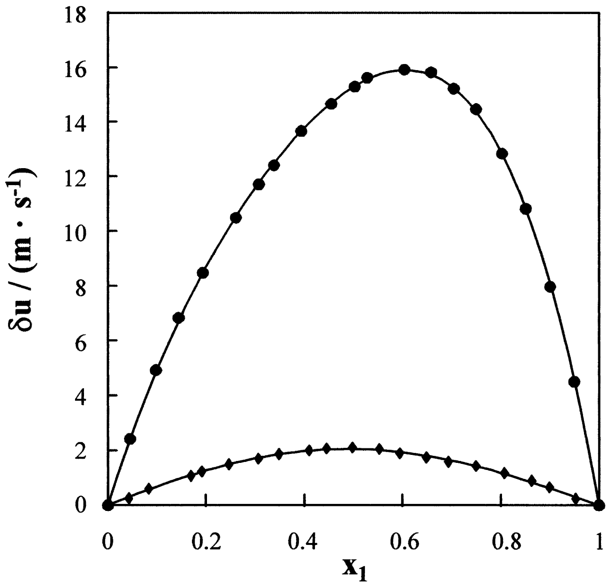

Figure 3. Change of speeds of sound on mixing of methanol (1) + ethyl butyrate (2) (●) and vinyl acetate (1) + ethyl butyrate (2) (◆) at 298.15 K.

Table 3. Adjustable Parameters a_k with the Standard Deviations σ for Excess Molar Volumes V^E, Refractive Index Deviations δn_D, and Speed of Sound Deviations δu

	$10^6 V^E$/m^3·mol^{-1}	δn_D	δu/m·s^{-1}
	Methanol (1) + Ethyl Butyrate (2)		
a_0	−0.0745	0.0689	61.1
a_1	−0.0020	−0.0399	−22.1
a_2	0.0806	0.0231	17.6
σ	8×10^{-10} (m^3·mol^{-1})	2×10^{-4}	6×10^{-2} (m·s^{-1})
	Vinyl Acetate (1) + Ethyl Butyrate (2)		
a_0	0.1901	−0.0017	8.1
a_1	−0.0279	0.0005	0.5
a_2	0.0358	0.0001	−1.5
σ	6×10^{-10} (m^3·mol^{-1})	3×10^{-5}	4×10^{-2} (m·s^{-1})

fitted to Redlich−Kister polynomials of the form

$$(V^E \text{ or } \delta n_D \text{ or } \delta u) = x_1 x_2 \sum_{k \geq 0} a_k (x_1 - x_2)^k \tag{4}$$

where a_k are the adjustable parameters obtained by a least-squares fit method and k is the degree of the polynomial expansion. Table 3 lists the parameters with the standard deviations σ. The coefficients a_k were used to calculate the solid curves in Figures 1−3. The standard deviations σ are defined as

$$\sigma = \sqrt{\frac{\sum (Z_{cal} - Z_{exp})_i^2}{N - m}} \tag{5}$$

where N is the number of experimental data, m is the number of equation parameters, and Z is the considered property (V^E or δn_D or δu).

VLE Data. Vapor−liquid equilibrium data (T, x_1, y_1) for the methanol (1) + ethyl butyrate (2) and vinyl acetate (1) + ethyl butyrate (2) binary systems at 101.3 kPa are presented in Table 4. The T−x_1−y_1 phase diagrams are ilustrated in Figures 4 and 5.

The fundamental equation of vapor−liquid equilibrium can be expressed by the equation

$$P\varphi_i y_i = P_i^s \varphi_i^s x_i \gamma_i \exp[V_i^L / RT(P - P_i^s)] \tag{6}$$

where P is the total pressure, x_i and y_i are the liquid- and vapor-phase mole fractions of component i, respectively, φ_i is the vapor-phase fugacity coefficient of component i in the mixture, and φ_i^s is the vapor-phase fugacity coefficient of pure component i at the system temperature and the corresponding satured vapor pressure P_i^s. V_i^L is the liquid molar volume of pure component i. At low pressure, the term $\exp[V_i^L/RT(P - P_i^s)]$ is approximately equal to 1. Then eq 6 may be simply rewritten for the vapor−liquid equilibrium as

$$P\varphi_i y_i = P_i^s \varphi_i^s x_i \gamma_i \tag{7}$$

The vapor-phase fugacity coefficients φ_i and φ_i^s are evaluated using the virial equation of state truncated after the second term. The second virial coefficients are calculated with the correlation of Tsonopoulos[7] and Prausnitz et al.[8] B_{12} was calculate with the Tsonopoulos modification for polar molecules by

$$B_{12} = \frac{RT_{c12}}{P_{c12}} (B^0 + \omega_{12} B^1 + a T_r^{-6} - b T_r^{-8}) \tag{8}$$

Table 4. Vapor−Liquid Equilibrium Data for the Methanol (1) + Ethyl Butyrate (2) and Vinyl Acetate (1) + Ethyl Butyrate (2) Systems: Liquid-Phase Mole Fraction (x_1), Vapor-Phase Mole Fraction (y_1), and Boiling Temperature (T), at 101.3 kPa

experimental data			Wilson equation			
T/K	x_1	y_1	T_{cal}/K	ΔT/K	$y_{1,cal}$	Δy_1
			Methanol (1) + Ethyl Butyrate			
382.3	0.042	0.332	380.2	2.1	0.374	−0.042
379.9	0.052	0.387	377.7	2.2	0.428	−0.041
374.6	0.074	0.487	372.9	1.7	0.519	−0.032
368.2	0.109	0.600	366.9	1.3	0.619	−0.019
360.8	0.162	0.709	360.3	0.5	0.713	−0.004
354.6	0.223	0.780	354.9	−0.3	0.778	0.002
350.6	0.292	0.835	350.6	0.0	0.823	0.012
349.8	0.307	0.844	349.9	−0.1	0.830	0.014
346.6	0.400	0.872	346.4	0.2	0.865	0.007
345.4	0.449	0.886	345.0	0.4	0.877	0.009
343.4	0.531	0.905	343.3	0.1	0.894	0.011
342.2	0.610	0.918	342.1	0.1	0.907	0.011
341.3	0.688	0.927	341.2	0.1	0.918	0.009
340.6	0.751	0.937	340.5	0.1	0.927	0.010
340.2	0.794	0.941	340.1	0.1	0.935	0.006
339.8	0.833	0.949	339.6	0.2	0.942	0.007
338.8	0.920	0.969	338.6	0.2	0.965	0.004
338.4	0.962	0.983	338.2	0.2	0.981	0.002
338.1	0.990	0.994	337.8	0.3	0.994	0.000
AAD[a]				0.5		0.013
			Vinyl Acetate (1) + Ethyl Butyrate			
392.0	0.025	0.087	392.2	−0.2	0.089	−0.002
387.9	0.069	0.217	388.2	−0.3	0.227	−0.010
384.5	0.114	0.327	384.3	0.2	0.346	−0.019
381.5	0.152	0.408	381.2	0.3	0.431	−0.023
378.8	0.192	0.480	378.2	0.6	0.508	−0.028
373.2	0.278	0.607	372.3	1.1	0.637	−0.030
370.9	0.324	0.662	369.5	1.4	0.691	−0.029
368.7	0.355	0.698	367.7	1.0	0.722	−0.024
366.6	0.396	0.736	365.5	0.9	0.758	−0.022
364.0	0.440	0.775	363.4	0.6	0.792	−0.017
362.0	0.487	0.808	361.3	0.7	0.822	−0.014
360.0	0.529	0.833	359.6	0.4	0.846	−0.013
358.3	0.572	0.857	358.0	0.3	0.868	−0.011
357.1	0.606	0.874	356.7	0.4	0.883	−0.009
354.4	0.679	0.906	354.3	0.1	0.912	−0.006
351.8	0.760	0.933	351.9	−0.1	0.938	−0.005
349.3	0.863	0.965	349.2	0.1	0.967	−0.002
348.1	0.914	0.979	347.9	0.2	0.980	−0.001
347.1	0.959	0.990	346.9	0.3	0.990	0.000
AAD[a]				0.5		0.003

[a] AAD = absolute average deviation.

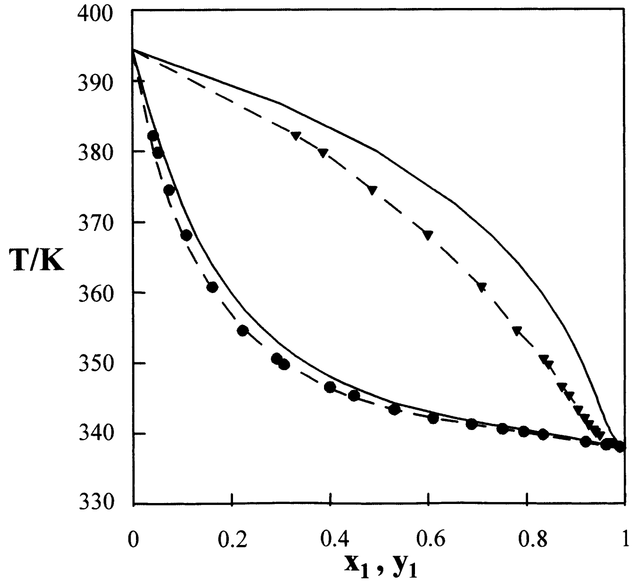

Figure 4. $T-x_1-y_1$ diagram for methanol (1) + ethyl butyrate (2) at 101.3 kPa: (●) experimental data of liquid-phase composition; (▲) experimental data of vapor-phase composition; (- - -) Wilson correlation; (—) ASOG prediction.

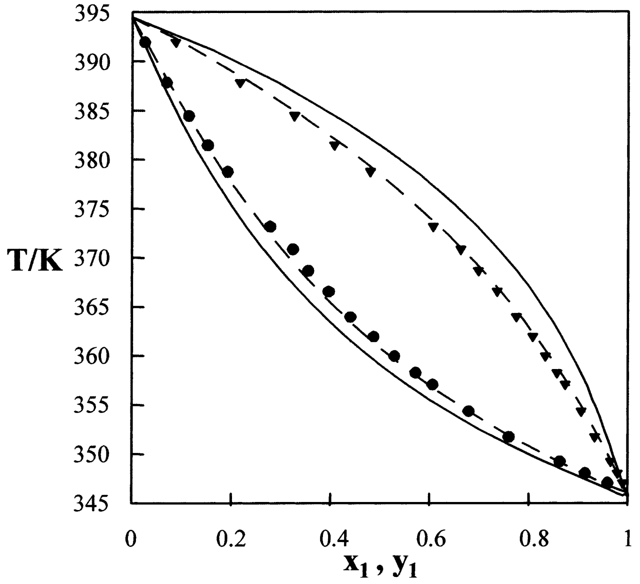

Figure 5. $T-x_1-y_1$ diagram for vinyl acetate (1) + ethyl butyrate (2) at 101.3 kPa: (●) experimental data of liquid-phase composition; (▲) experimental data of vapor-phase composition; (- - -) Wilson correlation; (—) ASOG prediction.

where a is the polarity parameter and b is the association parameter, T_r is the reduced temperature, and B^0 and B^1 are functions which depend exclusively on reduced temperature and can be represented satisfactorily by

$$B^0 = 0.083 - 0.422/T_r^{1.6} \tag{9}$$

$$B^1 = 0.139 - 0.172/T_r^{4.2} \tag{10}$$

The mixing rules proposed by Prausnitz[8] for the calculation of ω_{12}, T_{c12}, and P_{c12} are

$$\omega_{12} = \frac{\omega_1 + \omega_2}{2} \tag{11}$$

Table 5. Published Parameters[10] Used To Calculate the Fugacity Coefficients: Critical Temperature T_c, Critical Pressure P_c, Critical Volume V_c, Critical Compression Factor Z_c, and Acentric Factor ω of Pure Compounds

	T_c/K	$10^{-6}P_c$/Pa	V_c/m^3·kmol^{-1}	Z_c	ω
methanol	512.58	8.0959	0.117 80	0.224	0.5656
vinyl acetate	524.00	4.2500	0.270 00	0.263	0.3384
ethyl butyrate	571.00	3.0600	0.421 00	0.271	0.4190

Table 6. Coefficients of the Pure Substances for Eq 16

substance	A_i	B_i	C_i	D_i	E_i
methanol	109.93	−7471.3	−13.988	1.5281×10^{-2}	1
vinyl acetate	47.942	−5246.2	−3.636	4.5798×10^{-18}	6
ethyl butyrate	64.547	−6658.2	−6.0559	1.3960×10^{-17}	6

where ω_1 and ω_2 are the acentric factors of compounds 1 and 2, and

$$T_{c12} = (1 - k_{ij})(T_{c1}T_{c2})^{0.5} \tag{12}$$

where T_{c1} and T_{c2} are the critical temperatures of compounds 1 and 2, and k_{ij} is the binary interaction constant proposed by Lee and Chen,[9] for the alcohol + acetate mixtures, $k_{ij} = 0.08$.

Also,

$$P_{c12} = \frac{Z_{c12}RT_{c12}}{V_{c12}} \tag{13}$$

where Z_{c12} is calculated by

$$Z_{c12} = \frac{Z_{c1} + Z_{c2}}{2} \tag{14}$$

Z_{c1} and Z_{c2} are the critical compressibility factors, and V_{c12} is defined by the expression

$$V_{c12} = \left(\frac{V_{c1}^{1/3} + V_{c2}^{1/3}}{2}\right)^3 \tag{15}$$

where V_{c1} and V_{c2} are the critical volumes of compounds 1 and 2.

The critical properties P_c, V_c, T_c, and Z_c and the acentric factor ω have been obtained from the literature[10] and are presented in Table 5. The saturated vapor pressures were calculated with

$$\ln(P/\text{Pa}) = A_i + B_i/(T/\text{K}) + C_i \ln(T/\text{K}) + D_i(T/\text{K})^{E_i} \tag{16}$$

and the constants A_i, B_i, C_i, D_i, and E_i are reported in Table 6, and its values were obtained from the literature.[10]

The activity coefficients were correlated with the Margules, van Laar, Wilson,[11] NRTL,[12] and UNIQUAC[13] equations. To determine the constants of each model, we have used the method "VLE calc" suggested by Gess et al.[14] The parameters along with the average deviation in T (ΔT) and the average deviation in y (Δy) are listed in Table 7. Also, the ASOG[15] method was used to obtain the predictions in Figures 4 and 5.

The thermodynamic consistency of the experimental data was checked by means of a modified Dechema test.[16] According to this test, the experimental data are consistent if the average value for Δy is less than 0.01. The two systems included in this work have passed this consistency test; for methanol + ethyl butyrate, $\Delta y = 0.0088$, and for vinyl acetate + ethyl butyrate, $\Delta y = 0.0017$.

We also carried out the Margules constant test using the program of Gess et al.[14] The Margules constant can be used

Table 7. Correlation Parameters for Activity Coefficients, and Average Deviation for Studied Systems

equation	A_{12}	A_{21}	ΔT/K	Δy_1
Methanol (1) + Ethyl Butyrate (2)				
Margules[a]	0.7402	1.3432	0.3	0.011
van Laar[a]	0.8551	1.3883	0.6	0.014
Wilson[b] 2	4143.35	−23.16	0.5	0.013
NRTL[c] (α_{12} = 0.09)	8393.41	−3960.53	0.5	0.012
UNIQUAC[d]	−682.05	4791.26	0.5	0.012
Vinyl Acetate (1) + Ethyl Butyrate (2)				
Margules[a]	−0.0827	0.1284	0.3	0.003
van Laar[a]	0.0233	0.5237	0.3	0.003
Wilson[b] 2	−1485.75	3927.95	0.5	0.003
NRTL[c] (α_{12} = 0.45)	4398.59	−2526.04	0.2	0.003
UNIQUAC[d]	2829.48	−1833.15	0.2	0.004

[a] Margules and van Laar constants (dimensionless). [b] Wilson's interaction parameters (J·mol^{-1}). [c] NRTL's interaction parameters (J·mol^{-1}). [d] UNIQUAC's interaction parameters (J·mol^{-1}).

Table 8. Results of the Margules Constant Test

system	Margules constant
methanol (1) + ethyl butyrate (2)	0.9030
vinyl acetate (1) + ethyl butyrate (2)	0.0916

to indicate the ideality of a system. Systems which yield a Margules constant whose absolute value is less than 0.60 can be considered ideal, while those which yield an absolute value greater than 0.60 can be considered nonideal. This criterion for classification, however, is not rigorous. Table 8 shows the values of this constant.

Binary systems formed by methanol + ethyl butyrate and vinyl acetate + ethyl butyrate have no azeotrope, and the ethyl butyrate is feasible as an entrainer to break the azeotropic mixture formed by methanol and vinyl acetate. The ASOG method of prediction is not in good agreement with experimental data. For the binary system vinyl acetate + ethyl butyrate, the behavior is ideal, and the methanol + ethyl butyrate behavior is nonideal. For methanol + ethyl butyrate, the better equation of correlation for activity coefficients is the Margules equation, with ΔT = 0.3 K and Δy_1 = 0.011. For the system vinyl acetate + ethyl butyrate, ΔT and Δy_1 are very similar for all equations.The values of the excess molar volumes are very close to zero, especially for the methanol + ethyl butyrate system. The opposite behavior is observed for the changes of refractive index and speeds of sound on mixing.

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