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Isobaric Vapor—Liquid Equilibium for the Binary Systems of Methyl Formate with *o*-Xylene, *m*-Xylene, *p*-Xylene, and Ethylbenzene at 101.33 kPa

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ABSTRACT: Isobaric vapor—liquid equilibrium (VLE) data are reported for binary mixtures of methyl formate + *o*-xylene, methyl formate + *m*-xylene, methyl formate + *p*-xylene, and methyl formate + ethylbenzene at 101.33 kPa. The data are obtained using a vapor recirculating type (modified Othmer's) equilibrium still. All of the binary systems show positive deviation from ideality. None of the systems form an azeotrope. The VLE data for these binary systems are checked to meet rigorous thermodynamic consistency by the Herington method and the point-to-point test of the Fredenslund method. The experimental VLE data are well-correlated by the nonrandom two-liquid (NRTL), universal quasichemical activity coefficient (UNIQUAC), and Wilson equations.



■ INTRODUCTION

Ethylbenzene and xylenes are C₈ benzene homologues with the same molecular formula C₈H₁₀. The term of "mixed xylenes" describes a mixture of o-xylene, m-xylene, p-xylene, and ethylbenzene which are largely derived from petroleum. Mixed xylenes are generally a part of the commercial mixtures of solvent and gasoline, and the utilization rate of mixed xylenes is very low. As we know, these substances are of great importance in the petrochemical industry because they are the main basis for the synthesis of many organic compounds. For example, o-xylene is mainly oxidized to phthalic anhydride which is made into plasticizers, *m*-xylene is used to obtain isophthalic acid for use in polyesters, p-xylene is converted to terephthalic acid or to dimethyl terephthalate as the first stage in the production of polyester, and ethylbenzene is dehydrogenated to styrene. The production of individual components of very high purity from C₈ aromatic isomers by distillation is difficult due to the low relative volatilities among these compounds.

Berg¹ has reported that methyl formate can be used as an agent for the separation of C_8 aromatic isomers. Unfortunately, no vapor—liquid equilibria (VLE) data for the binary systems of (methyl formate + ethylbenzene, or *o*-, or *m*-, or *p*-xylene) have been reported in the literature at either isobaric or isothermal conditions. However, it is known that accurate VLE data is essential in the simulation and design of the distillation process. Isobaric VLE data at 100.65 kPa for six binary systems of C_8 compounds are published in the literature.^{2,3}

In this work, the isobaric VLE data of the four binary systems methyl formate + o-, or m-, or p-xylene, or ethylbenzene, which have wide boiling points, are determined at a pressure of 101.33 kPa. The thermodynamic consistency are tested for these four systems. The nonrandom two-liquid (NRTL),⁴ universal quasichemical activity coefficient (UNIQUAC),⁵ and Wilson⁶ equations are used to correlate the experimental data of each binary mixture for their correlation parameters.

EXPERIMENTAL SECTION

Materials. Methyl formate is supplied by Sinopharm Chemical Reagent Co., Ltd. with a minimum mass fraction purity of 0.980. Ethylbenzene is provided by Sinopharm Chemical Reagent Co., Ltd. and has a minimum mass fraction purity of 0.985. The o-xylene, m-xylene, and p-xylene, which are all analytical reagent (AR) grade materials with a minimum mass fraction purity of 0.990, are bought from Aladdin Chemistry Co., Ltd. All chemicals are dried over 0.4 nm molecular sieves and then distilled and degassed as described by Fischer and Gmehling⁷ before use. The purity of the materials is checked by gas chromatography and found to be greater than 0.998 mass fraction. The densities of the pure components are measured at a temperature of 298.15 K using an Anton Paar DMA 58 densimeter, with an accuracy of \pm 0.01 kg·m⁻³, and the refractive indices of the pure liquids are determined using a calibrated Abbe refractometer with an uncertainty of ± 0.0002 at 298.15 K. The temperature is controlled to \pm 0.01 K with a thermostat bath. The densities, refractive indices, and normal boiling points for these compounds are given in Table 1 together with their literature values. $^{\rm 8-10}$

Apparatus and Procedure. The VLE data are obtained by using a modified equilibrium still similar to Othmer still^{11,12} with some modifications described elsewhere.^{13–15} The modified equilibrium still and experimental procedure are described in detail in the previous work.¹⁶ This still can prevent the occurrence of violent boiling efficiently. It takes approximately 2 h for the circulation in the Othmer cell to establish VLE when temperature is kept constant. The samples of the liquid and condensed vapor are taken out for analysis.

The temperature is measured with the help of a standard mercury thermometer, and the uncertainty of temperature

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Table 1. Densities (ρ), Refractive Index ($n_{\rm D}$), and Boiling	Points of Pure Compounds Co	mpared with Literature Data
$p/(1-m^{-3})$ (209.15 K)	·· (200.15 K)	T / V (101.2 l-D)

	$\rho/(\text{kg}\cdot\text{m}^{-3})$ (298.15 K)		$n_{\rm D}$ (2)	n _D (298.15 К)		T _b /K (101.3 kPa)	
compound	exptl	lit.	exptl	lit.	exptl	lit.	
methyl formate	966.38	966.40 ^a	1.3412	1.34150 ^a	304.89	304.900 ^b	
o-xylene	875.85	875.90 ^c	1.5028	1.50295 ^c	417.56	417.579 ^c	
<i>m</i> -xylene	859.87	860.00 ^c	1.4942	1.49464 ^c	412.21	412.270 ^c	
<i>p</i> -xylene	856.46	856.70 ^c	1.4932	1.49325 ^c	411.37	411.509 ^c	
ethylbenzene	862.55	862.60 ^c	1.4930	1.49320 ^c	409.32	409.352 ^c	
Reference 8. ^b Reference 9. ^c Reference 10.							

measurements is \pm 0.1 K. The pressure is fixed and kept at (101.33 \pm 0.04) kPa by using an automatic pressure control system. It consists of one electromagnetic relay, one vacuum pump, two manometers, two reservoirs and three triple valves. The principle of the pressure control system is described in detail in our previous work.¹⁷

The liquid and condensed vapor samples are analyzed using a gas chromatograph (GC) SP6890A. A flame ionization detector (FID) is used together with a SE-54 capillary column (30 m \times 0.32 mm \times 1 μ m). High-purity nitrogen with a purity of 99.999 % is used as the carrier gas at a constant flow rate of 30 cm³·min⁻¹. The GC responses are treated with a dual-channel chromatography workstation N-2000 supplied by Zhejiang Zhida Information Engineering Co., Ltd. The column, injector, and detector temperatures are kept at (393, 433, and 433) K, respectively. The GC is calibrated with a set of solutions of known compositions that are prepared gravimetrically by an electronic analytic balance (with an uncertainty of \pm 0.0001 g). A single analysis of the vapor or liquid composition by GC is frequently imprecise. At least three analyses are made for each sample. The standard uncertainty of the measured mole fraction is \pm 0.001.

RESULTS AND DISCUSSION

The isobaric VLE data and the calculated activity coefficients are listed in Tables 2 to 5 for (methyl formate + *o*-xylene),

Table 2. Experimental	Vapor-Liquid	Equilibrium	Data for
Methyl Formate (1) +	o-Xylene (2) a	t 101.33 kPa	

T/K	x_1	y_1	γ_1	γ_2
305.99	0.9282	0.9969	1.0326	4.5476
306.91	0.8566	0.9956	1.0799	3.0226
307.47	0.7597	0.9950	1.1921	2.0823
307.85	0.7042	0.9947	1.2678	1.7668
309.32	0.5958	0.9913	1.4153	1.3069
312.96	0.4184	0.9885	1.7640	1.2838
318.45	0.3227	0.9769	1.8690	1.0373
328.66	0.1957	0.9667	2.1840	1.0307
345.70	0.1038	0.9139	2.3451	1.0278
362.49	0.0584	0.8356	2.3468	1.0236
378.59	0.0327	0.6858	2.3997	1.0179
394.72	0.0157	0.4701	2.4284	1.0038
397.29	0.0134	0.4295	2.4362	1.0015
403.20	0.0089	0.3122	2.4387	0.9930
406.53	0.0071	0.2602	2.4723	0.9882
411.14	0.0037	0.1581	2.4840	0.9858
412.92	0.0028	0.1229	2.5019	0.9848

(methyl formate + m-xylene), (methyl formate + p-xylene), and (methyl formate + ethylbenzene), respectively.

Table 3. Experim	ental Vapor–Liquid Equilibrium Dat	a for
Methyl Formate	(1) + <i>m</i> -Xylene (2) at 101.33 kPa	

T/K	x_1	<i>y</i> ₁	γ_1	γ_2
305.66	0.9710	0.9973	0.9997	4.8027
306.43	0.9296	0.9943	1.0117	3.5876
306.99	0.8977	0.9925	1.0243	2.8231
308.03	0.8238	0.9897	1.0712	2.0975
309.01	0.7216	0.9874	1.1771	1.5379
309.49	0.6726	0.9866	1.2400	1.3884
310.04	0.6090	0.9857	1.3413	1.2518
311.34	0.5749	0.9852	1.3553	1.2002
315.57	0.4670	0.9833	1.4349	1.1097
317.62	0.4234	0.9821	1.4731	1.1061
321.90	0.3474	0.9795	1.5507	1.0243
332.07	0.2275	0.9468	1.6546	1.0085
337.68	0.1888	0.9384	1.6682	1.0008
345.83	0.1409	0.9084	1.7110	0.9998
357.50	0.0940	0.8367	1.7238	0.9975
364.28	0.0738	0.7892	1.7427	0.9964
372.88	0.0537	0.7184	1.7676	0.9964
379.77	0.0403	0.6320	1.7692	0.9963
388.20	0.0268	0.5171	1.7883	0.9958
399.05	0.0134	0.3221	1.8037	0.9947
411.07	0.0009	0.0323	2.1042	0.9936

Table 4. Experimental	Vapor–Liquid Equilibrium Data for
Methyl Formate (1) +	<i>p</i> -Xylene (2) at 101.33 kPa

T/K	x_1	y_1	γ_1	γ_2
306.10	0.9414	0.9959	1.0129	3.8875
306.65	0.9128	0.9945	1.0221	3.8161
307.24	0.8795	0.9912	1.0345	3.4020
308.50	0.7882	0.9892	1.0999	2.4913
309.36	0.7093	0.9874	1.1824	2.0234
309.52	0.6963	0.9871	1.1971	1.9663
310.32	0.6202	0.9857	1.3039	1.6712
311.37	0.5533	0.9843	1.4054	1.4769
314.87	0.3744	0.9789	1.8256	1.2039
317.87	0.2978	0.9742	2.0599	1.1846
319.44	0.2747	0.9688	2.1057	1.1107
342.47	0.0985	0.9091	2.6942	1.0198
358.19	0.0546	0.8330	2.8296	1.0168
364.04	0.0440	0.7759	2.8749	1.0126
374.24	0.0302	0.6779	2.8910	1.0022
389.11	0.0146	0.4700	2.9022	0.9995
394.44	0.0109	0.3934	2.9506	0.9991
402.18	0.0057	0.2310	2.9538	0.9982

The activity coefficients of the components in the liquid phase are calculated using the equations¹⁸ expressed as follows:

$$\gamma_1 = (py_1/p_1^0 x_1) \exp[\{(B_{11} - V_1)(p - p_1^0)/RT\} + (p\delta_{12}y_2^2)/RT]$$
(1)

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Table 5. Experimental Vapor-Liquid Equilibrium Data for Methyl Formate (1) + Ethylbenzene (2) at 101.33 kPa

T/K	x_1	y_1	γ_1	γ_2
305.70	0.9665	0.9975	1.0031	3.8879
306.27	0.9397	0.9958	1.0083	3.2373
306.41	0.9072	0.9944	1.0375	2.6983
306.61	0.8814	0.9924	1.0579	2.3736
307.02	0.8492	0.9902	1.0790	2.0566
307.66	0.7657	0.9895	1.1680	1.5723
309.43	0.6758	0.9868	1.2371	1.3092
312.33	0.5753	0.9864	1.3062	1.1771
314.47	0.5336	0.9844	1.3090	1.1572
318.41	0.4425	0.9762	1.3414	1.1027
321.71	0.3705	0.9718	1.3638	1.0565
328.37	0.2967	0.9638	1.4101	1.0353
343.44	0.1778	0.9019	1.4184	1.0305
355.69	0.1190	0.8400	1.4188	1.0185
364.08	0.0895	0.7874	1.4223	0.9997
370.68	0.0698	0.7104	1.4333	0.9958
379.06	0.0500	0.6208	1.4402	0.9948
387.52	0.0311	0.4651	1.4409	0.9945
395.33	0.0197	0.3458	1.4494	0.9938
401.32	0.0101	0.1907	1.4517	0.9932

$$\gamma_2 = (py_2/p_2^0 x_2) \exp[\{(B_{22} - V_2)(p - p_2^0)/RT\} + (p\delta_{12}y_1^2)/RT]$$
(2)

where

$$\delta_{12} = 2B_{12} - B_{11} - B_{22} \tag{3}$$

where *P* is the total pressure; *T* is the temperature; x_1 , x_2 and y_1 , y_2 are the mole fractions of components 1 and 2 in the liquid and vapor phases, respectively; V_1 and V_2 are the liquid molar volumes of pure components 1 and 2, calculated using the Yen and Woods¹⁹ method; *R* is the gas constant; B_{11} and B_{22} are the second virial coefficients of the pure components; B_{12} is the cross second virial coefficient; and p_1^0 and p_2^0 are the vapor pressure of pure components 1 and 2. The pure component vapor pressures (p^0) are calculated using the Antoine equation

$$\log p^0 / k Pa = A - B / (C + t/^{\circ}C)$$
⁽⁴⁾

where A, B, and C are Antoine constants and their values⁹ are reported in Table 6.

The Pitzer and Curl equation modified by Tsonopoulos²⁰ is used for the estimation of second virial coefficients as well as cross second virial coefficients in this work. The physical properties⁹ of the pure components needed in this calculation are also shown in Table 6.

Table 6. Physical Properties of the Pure Compounds^a

Article

The $T-x_1-y_1$ plots for the binary systems of (methyl formate + o-, or m-, or p-xylene or ethylbenzene) at 101.33 kPa are given in Figures 1 to 4, respectively. The four binary systems do



Figure 1. $T-x_1-y_1$ diagram for methyl formate (1) + *o*-xylene (2) at 101.33 kPa: \bullet , experimental liquid-phase mole fractions, x_1 ; \bigcirc , experimental vapor-phase mole fractions, y_1 ; —, NRTL equation.



Figure 2. $T-x_1-y_1$ diagram for methyl formate (1) + *m*-xylene (2) at 101.33 kPa: \bullet , experimental liquid-phase mole fractions, x_1 ; O, experimental vapor-phase mole fractions, y_1 ; —, NRTL equation.

not present azeotropes at 101.33 kPa as shown in Figures 1 to 4. Figures 5 to 8 show the relationships between activity coefficients γ_1 and γ_2 and mole fractions x_1 , we can observe that all of the binary systems show positive deviations from ideal behavior.

	methyl formate	o-xylene	<i>m</i> -xylene	<i>p</i> -xylene	ethylbenzene
T_{c}^{b}/K	487.2	630.2	617.0	616.2	617.1
P _c ^b ∕MPa	5.998	3.729	3.546	3.516	3.607
$V_{c}^{b}/cm^{3}\cdot mol^{-1}$	172.0	369.0	376.0	379.0	374.0
μ^b/D	1.8	1.5	0.3	0.1	0.4
ω^b	0.252	0.314	0.331	0.324	0.301
		Constants of Antoine'	s Equation ^b , eq 4		
Α	6.29530	6.12381	6.13398	6.11542	6.08208
В	1125.200	1474.679	1462.266	1453.430	1424.255
С	230.560	213.686	215.105	215.307	213.060

^aCritical temperature, T_{ci} critical pressure, P_{ci} critical volume, V_{ci} dipole moment, μ_i acentric factor, ω . ^bReference 9.



Figure 3. $T-x_1-y_1$ diagram for methyl formate (1) + *p*-xylene (2) at 101.33 kPa: \bullet , experimental liquid-phase mole fractions, x_1 ; \bigcirc , experimental vapor-phase mole fractions, y_1 ; -, NRTL equation.



Figure 4. $T-x_1-y_1$ diagram for methyl formate (1) + ethylbenzene (2) at 101.33 kPa: •, experimental liquid-phase mole fractions, x_1 ; O, experimental vapor-phase mole fractions, y_1 ; -, NRTL equation.



Figure 5. Plot of γ_1 and γ_2 vs x_1 for the system methyl formate (1) + *o*-xylene (2) at 101.33 kPa: \blacksquare , γ_1 and O, γ_2 , experimental data; —, calculated data using the NRTL equation.

The four systems are thermodynamically consistent according to the Herington method²¹ because the check result D–J is



Figure 6. Plot of γ_1 and γ_2 vs x_1 for the system methyl formate (1) + *m*-xylene (2) at 101.33 kPa: \blacksquare , γ_1 and \bigcirc , γ_2 , experimental data; —, calculated data using the NRTL equation.



Figure 7. Plot of γ_1 and γ_2 vs x_1 for the system methyl formate (1) + *p*-xylene (2) at 101.33 kPa: \blacksquare , γ_1 and O, γ_2 , experimental data; —, calculated data using the NRTL equation.



Figure 8. Plot of γ_1 and γ_2 vs x_1 for the system methyl formate (1) + ethylbenzene (2) at 101.33 kPa: \blacksquare , γ_1 and O, γ_2 , experimental data; —, calculated data using the NRTL equation.

less than 10. In this work, the values of D–J for methyl formate + p-xylene, methyl formate + m-xylene, methyl formate + p-xylene,

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and methyl formate + ethylbenzene systems are -28.56, -30.94, -43.29, and -26.27, respectively.



Figure 9. Results of the thermodynamic consistency test for VLE data of the methyl formate (1) + *o*-xylene (2) system using the Fredenslund method. \blacksquare , $y_{1,cal} - y_{1,exp}$; O, $P_{cal} - P_{exp}$.



Figure 10. Results of the thermodynamic consistency test for VLE data of the methyl formate (1) + *m*-xylene (2) system using the Fredenslund method. \blacksquare , $y_{1,cal} - y_{1,exp}$; O, $P_{cal} - P_{exp}$.



Figure 11. Results of the thermodynamic consistency test for VLE data of the methyl formate (1) + *p*-xylene (2) system using the Fredenslund method. \blacksquare , $y_{1,eal} - y_{1,exp}$; O, $P_{cal} - P_{exp}$.

The experimental data of the binary systems are tested for thermodynamic consistency by means of the point-to-point test of Van Ness,²² modified by Fredenslund et al.²³ According to



Figure 12. Results of the thermodynamic consistency test for VLE data of the methyl formate (1) + ethylbenzene (2) system using the Fredenslund method. \blacksquare , $y_{1,cal} - y_{1,exp}$; O, $P_{cal} - P_{exp}$.

Table 7. Interaction Parameters and the Root-Mean-Squared Deviations between Calculated and Experimental Vapor-Phase Mole Fractions σy_1 and Temperature $\sigma T/K$ for the Binary Systems with Different Models

	moo	lel parameters				
	A_{12}^{a} A_{21}^{a}			root-mear devia	n-squared ition	
model	J·mol⁻¹	J·mol⁻¹	α_{12}	$\sigma y_1^{\ b}$	$\sigma T^b/K$	
	Methyl Fo	ormate $(1) + o$	-Xylene (2)		
NRTL	5147.45	-506.07	0.30	0.0040	0.50	
UNIQUAC	-585.80	-843.37		0.0066	0.74	
Wilson	-1570.76	-2134.54		0.0057	0.84	
	Methyl Fo	rmate (1) + <i>m</i>	-Xylene (2	.)		
NRTL	6300.43	-1499.68	0.30	0.0040	0.58	
UNIQUAC	-1506.25	258.15		0.0046	0.62	
Wilson	552.63	-5514.18		0.0064	0.73	
	Methyl Fo	ormate $(1) + p$	-Xylene (2)		
NRTL	3441.08	1016.22	0.30	0.0034	0.28	
UNIQUAC	184.32	-1882.29		0.0046	0.39	
Wilson	-1416.62	-3560.22		0.0034	0.35	
	Methyl Forn	nate (1) + Eth	ylbenzene	(2)		
NRTL	7797.70	-109.25	0.30	0.0043	0.55	
UNIQUAC	-2129.55	849.69		0.0147	1.18	
Wilson	1170.28	-5826.45		0.0114	0.95	
The interaction nerror store for versions models are as fulles NDTI						

^{...}The interaction parameters for various models are as follows: NRTL: $A_{12} = (g_{12} - g_{22}), A_{21} = (g_{21} - g_{11});$ UNIQUAC: $A_{12} = (U_{12} - U_{22}),$ $A_{21} = (U_{21} - U_{11});$ Wilson: $A_{12} = (\lambda_{12} - \lambda_{11}), A_{21} = (\lambda_{21} - \lambda_{22}).$ ^b $\sigma T = [\sum_{i=1}^{n} (T_{i}^{cal} - T_{i}^{exp})^{2}/n]^{1/2}; \sigma y_{1} = [\sum_{i=1}^{n} (y_{1,i}^{cal} - y_{1,i}^{exp})^{2}/n]^{1/2}.$

this test, a set of experimental data are considered consistent if the mean absolute deviation between calculated and measured vapor phase compositions is less than 0.01. Moreover, if the pressure residuals are all smaller than 10 mmHg (1.33 kPa), the experimental data are also considered thermodynamically consistent. The results of this test for the four binary systems are given in Figures 9 to 12, and the mean absolute deviations in the vapor phase mole fraction are 0.0033, 0.0038, 0.0020, and 0.0033 for methyl formate + o-xylene, methyl formate + m-xylene, methyl formate + p-xylene, and methyl formate + ethylbenzene, respectively, indicating that the experimental data of all binary systems pass the Fredeslund consistency test.

The VLE experimental data are correlated with NRTL, UNIQUAC, and Wilson equations by minimizing the objective



Figure 13. Experimental and calculated $T-y_1$ diagram for methyl formate (1) + *o*-xylene (2) at 101.33 kPa: \blacksquare , $T-y_1$ experimental; \bigcirc , $T-y_1$ calculated (calculation based on the NRTL model); \triangle , $T-y_1$ calculated (calculation based on the UNIQUAC model); \Diamond , $T-y_1$ calculated (calculation based on the Wilson model).



Figure 14. Experimental and calculated $T-y_1$ diagram for methyl formate (1) + *m*-xylene (2) at 101.33 kPa: \blacksquare , $T-y_1$ experimental; \bigcirc , $T-y_1$ calculated (calculation based on the NRTL model); \triangle , $T-y_1$ calculated (calculation based on the UNIQUAC model); \Diamond , $T-y_1$ calculated (calculation based on the Wilson model).

function F in terms of calculated and experimental activity coefficient values.²⁴ The function F used in this work can be expressed as

$$F = \sum_{k=1}^{N} \{ (\gamma_1^{\exp} - \gamma_1^{cal})_k^2 + (\gamma_2^{\exp} - \gamma_2^{cal})_k^2 \}$$
(5)

where γ is the activity coefficient and *N* denotes the number of experimental data.

The correlation deviations of the vapor phase composition and equilibrium temperature for each data point can be seen in Figures 13 to 16. The interaction parameters A_{12} and A_{21} for the NRTL, UNIQUAC, and Wilson equations along with the root-mean-squared deviations (rmsd) between the experimental and calculated values of equilibrium temperature and vapor phase mole fraction are listed in Table 7. According to comparisons between experimental data and calculated data displayed in Figures 13 to 16, it can be observed that the



Figure 15. Experimental and calculated $T-y_1$ diagram for methyl formate (1) + *p*-xylene (2) at 101.33 kPa: \blacksquare , $T-y_1$ experimental; \bigcirc , $T-y_1$ calculated (calculation based on the NRTL model); \triangle , $T-y_1$ calculated (calculation based on the UNIQUAC model); \Diamond , $T-y_1$ calculated (calculation based on the Wilson model).



Figure 16. Experimental and calculated $T-y_1$ diagram for methyl formate (1) + ethylbenzene (2) at 101.33 kPa: \blacksquare , $T-y_1$ experimental; \bigcirc , $T-y_1$ calculated (calculation based on the NRTL model); \triangle , $T-y_1$ calculated (calculation based on the UNIQUAC model); \diamondsuit , $T-y_1$ calculated (calculation based on the Wilson model).

experimental data agree well with data calculated by using NRTL, UNIQUAC, and Wilson models.

CONCLUSIONS

The isobaric VLE data for methyl formate + *o*-xylene, methyl formate + *m*-xylene, methyl formate + *p*-xylene, and methyl formate + ethylbenzene are obtained using a modified Othmer cell at 101.33 kPa. No azeotropes are observed. The VLE data of all of the measured binary sytems are tested for thermodynamic consistency on the basis of Herrington method and Fredenslund method and found to be consistent. The experimental data are correlated with three activity coefficient models, namely, NRTL, UNIQUAC, and Wilson. The correlated results are in good agreement with experimental data, which indicates that all equations are suitable for correlating the data for the mixtures considered here.

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