Isobaric Vapor−Liquid Equilibrium for the Ternary System *o*-Xylene + Nonane + Cyclohexanol

Enn Siimer,* Helle Kirss, Mati Kuus, and Ludmilla Kudryavtseva

Institute of Chemistry at Tallinn Technical University, Akadeemia tee 15, 12618 Tallinn, Estonia

Isobaric vapor−liquid equilibria (VLE) were measured for the system *o*-xylene + nonane + cyclohexanol and for two constituent binaries containing cyclohexanol at the pressures 26.66, 53.33, and 79.99 kPa. The new modified Wilson equation is proposed to better describe vapor−liquid equilibria (VLE) in binary and multicomponent systems. The equation takes into account the linear dependence of the difference of interaction energies $\lambda_{ij} - \lambda_{ii}$ on temperature and avoids the use of molar volumes. Correlation of binary isobaric VLE experimental data gave four coefficients that can describe VLE at different pressures and in a large boiling temperature region. It was found that VLE data for the ternary system *o*-xylene + nonane + cyclohexanol can be predicted by the modified Wilson equation.

Introduction

Among two-parameter equations (Margules, van Laar, UNIQUAQ, UNIFAC), the Wilson equation[1] is widely used. In the introduction of the Thermodynamics Data Center (Poland) collection,[2] it has been confirmed that for alcohol + aliphatic hydrocarbon mixtures the Wilson equation gives the best results.

For description of vapor−liquid equilibria (VLE) binary isobaric experimental data, two Wilson parameters Λ_{ij} are valid for an average boiling temperature T interval used:

$$\Lambda_{ij} = \frac{V_j}{V_i} \exp\left[-\frac{\lambda_{ij} - \lambda_{ii}}{RT}\right] \quad (1)$$

where V_i and V_j are the molar volumes of components i and j.

The other possibility is to calculate the $\lambda_{ij} - \lambda_{ii}$ values in the correlation procedure instead of Λ_{ij}. In this case, for a binary system, $\lambda_{12} - \lambda_{11}$ and $\lambda_{21} - \lambda_{22}$ are regarded as constant parameters for VLE description.

In our previous paper[3] we have shown that it is reasonable to consider the dependence of the parameter $(\lambda_{ij} - \lambda_{ii})$ on temperature, as proposed earlier.[4] The next equations for binary systems are given by

$$\Lambda_{12} = \frac{V_2}{V_1} \exp\left[-\frac{(m_1 + n_1 T)}{RT}\right] \quad (2)$$

$$\Lambda_{21} = \frac{V_1}{V_2} \exp\left[-\frac{(m_2 + n_2 T)}{RT}\right] \quad (3)$$

where T is an actual boiling temperature measured at a chosen pressure and molar composition.

In an earlier paper,[3] the VLE calculation, by means of these relationships, was successfully applied to the strongly nonideal system 1-nonyne + 1-propanol at five pressures.

In this article, we present simplified equations for Λ_{ij} instead of eqs 2 and 3. These equations can describe VLE on the basis of isobaric or isothermal experimental data.

The vapor−liquid equilibrium for the ternary system *o*-xylene + nonane + cyclohexanol was measured in this work, and for the obtained data, the new equations were tested. Isobaric VLE data at three pressures (26.66, 53.33, and 79.99 kPa) are presented for this system and for two binaries containing cyclohexanol. No VLE data were found for these systems in the literature. The vapor−liquid equilibium for *o*-xylene + nonane has been measured by us earlier.[5] One set of isobaric data at the pressure 101.32 kPa for the system *o*-xylene + nonane is available.[6]

Azeotropic parameters were determined for the ternary system and two constituent binaries *o*-xylene + cyclohexanol and nonane + cyclohexanol. Those of *o*-xylene + nonane have been reported earlier.[5]

Experimental Section

Materials. *o*-Xylene and nonane were of the same purity, greater than 99.5 and 99.8 mass %, respectively, as reported in our earlier work.[5] Cyclohexanol was dried with fresh ignited CaO and distilled at a subatmospheric pressure in a high-efficiency Teflon rotor-column. The final purity of cyclohexanol determined by GLC (flame ionization detector, glass tube column with Chromaton N-AW-DMCS and 5% DC550) was greater than 99.5 mass %. In Table 1, the densities measured with a capillary pycnometer, refractive indices measured with a calibrated Abbé refractometer, and boiling temperatures are compared with those given in the literature.

Apparatus and Procedure. The $T-x$ results were obtained by the semi-microebulliometer described by us in detail.[5,7] Uncertainties of the boiling temperature were estimated to be less than 0.05 K. The amount of mixtures investigated was less than 1 mL. Uncertainties of mole fraction composition of a liquid mixture (x) were estimated to be less than 5×10^{-4}.

Modified Expressions for Wilson Parameters

By setting in eqs 2 and 3

$$a_{ij} = -\frac{n_i}{R} + \ln\frac{V_j}{V_i} \quad (4)$$

$$b_{ij} = -\frac{m_i}{R} \quad (5)$$

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Table 1. Densities *d*, Refractive Indices n_D, and Normal Boiling Points T_b of Pure Components

	d/kg·m^{-3}			n_D			T_b/K	
component	T/K	obs	lit.	T/K	obs	lit.	obs	lit.
o-xylene	293.15	880.1	880.1[a]	293.15	1.5054	1.505 45[a]	417.53	417.55[b]
nonane	293.15	717.6	717.72[a]	293.15	1.4055	1.405 42[a]	423.97	423.95[b]
cyclohexanol	303.15	941.6	941.6[b]	303.15	1.4621	1.462 9[b]	433.89	434.25[b]

[a] *TRC Thermodynamic Tables*.[8] [b] Dean.[9]

Table 2. Isobaric Vapor−Liquid Equilibrium Data: Liquid-Phase Mole Fraction x_1 and Boiling Temperatures T in Binary Systems

	T/K at pressure P/kPa		
x_1	$P = 26.66$	$P = 53.33$	$P = 79.99$
	o-Xylene (1) + Cyclohexanol (2)		
0.000	394.86	413.79	426.16
0.173	384.71	405.21	418.44
0.184	385.09	405.36	418.52
0.260	381.67	402.44	415.91
0.332	379.59	400.51	414.05
0.421	377.72	398.73	412.35
0.508	376.37	397.40	411.05
0.596	375.36	396.45	410.15
0.614	375.04	396.14	409.85
0.697	374.28	395.38	409.13
0.797	373.55	394.78	408.60
0.898	373.15	394.50	408.42
1.000	373.40	394.86	408.82
	Nonane (1) + Cyclohexanol (2)		
0.000	394.86	413.79	426.16
0.119	384.75	404.99	418.06
0.225	380.56	401.04	414.27
0.317	378.75	399.18	412.44
0.427	377.75	398.10	411.37
0.503	377.14	397.52	410.82
0.600	376.54	397.04	410.46
0.651	376.44	396.90	410.29
0.704	376.40	396.93	410.39
0.749	376.37	397.05	410.62
0.835	376.48	397.36	411.08
0.920	377.47	398.74	412.70
1.000	379.88	401.28	415.26

Table 3. Fitted Coefficients of the Modified Wilson Equation (Eqs 7 and 8) and Calculated Absolute Mean Errors (Δ*P*) and Standard Deviations (σ*P*) of Pressure for Binary Systems

system (1) + (2)	a_{12}	b_{12}	a_{21}	b_{21}	ΔP/%	σP/kPa
o-xylene + cyclohexanol	0.212 56	−129.163	0.212 56	−129.163	0.294	0.22
nonane + cyclohexanol	0.056 76	−314.903	3.585 44	−1774.377	0.203	0.15
o-xylene + nonane	0.143 82	41.904	0.231 69	−315.517	0.094	0.08

Table 4. Coefficients for the Antoine Vapor Pressure Equation (kPa, K) (Eq 12)

component	A_i	B_i	C_i	temp region
o-xylene	14.043 69	3352.595	−61.832	(373 to 418) K
nonane	13.854 59	3224.816	−74.824	(380 to 424) K
cyclohexanol	13.721 85	2778.058	−128.7243	(395 to 426) K

Table 5. Azeotropic Boiling Temperatures T_{az} and Compositions x_1^{az} of Binary Systems versus Pressure *P*

	o-xylene (1) + cyclohexanol (2)		nonane (1) + cyclohexanol (2)	
P/kPa	T_{az}/K	x_1^{az}	T_{az}/K	x_1^{az}
26.66	373.1	0.945	376.3	0.742
53.33	394.4	0.905	396.9	0.695
79.99	408.3	0.875	410.2	0.665

we obtain

$$\Lambda_{ij} = \exp\left[a_{ij} + \frac{b_{ij}}{T}\right] \tag{6}$$

and for binary systems

$$\Lambda_{12} = \exp\left[a_{12} + \frac{b_{12}}{T}\right] \tag{7}$$

$$\Lambda_{21} = \exp\left[a_{21} + \frac{b_{21}}{T}\right] \tag{8}$$

As seen from eqs 4 and 5, the parameters a_{12}, b_{12}, a_{21}, and b_{21} simplify calculation procedures, because they include molar volumes V_1 and V_2 and the gas constant R. The ratio of molar volumes V_j/V_i usually does not depend considerably on temperature, but eqs 7 and 8 take into account its temperature dependence.

Consequently, the Wilson equation with two parameters at a given pressure could be replaced with a four-parameter equation. These parameters describe VLE in a large boiling temperature region for simultaneous use at different chosen pressures in isobaric experiments. This means that actually the number of required parameters decreases. For example, as we have shown, VLE of the system 1-nonyne + 1-propanol system can be described with four parameters at five pressures in a large temperature range from 324.98 to 424.21 K.[3]

For practical calculation of the parameters a_{12}, b_{12}, a_{21}, and b_{21} in the correlation procedure, different iteration methods can be used. As we have demonstrated,[3] the Newton iteration method is recommended because analytical expressions of partial derivatives of the first and second order are easy to find. Convergence is rapid, and the set of initial approximations 0, 0, 0, 0 or 1, 0, 1, 0 can be used.

The prediction of VLE in multicomponent systems can be realized on the basis of binary parameters. For example, the values of activity coefficients in ternary systems can be expressed as

$$\gamma_i = \exp\left[1 - \ln\left(\sum_{j=1}^{3} x_j\Lambda_{ij}\right) - \sum_{k=1}^{3}\frac{x_k\Lambda_{ki}}{\sum_{j=1}^{3} x_j\Lambda_{kj}}\right] \tag{9}$$

and the Gibbs energy G^E as

$$G^E = -RT\left(\sum_{i=1}^{3} x_i \ln\sum_{j=1}^{3} x_j\Lambda_{ij}\right) \tag{10}$$

where x_i is the mole fraction of component i.

For predicting VLE in ternary systems, values of 12 parameters must be previously found for the binaries.

Table 6. Experimental Vapor−Liquid Equilibria Data for the Ternary System *o*-Xylene (1) + Nonane (2) + Cyclohexanol (3) at Three Pressures and Values Calculated by Using Eqs 7 and 8 of Activity Coefficients γ_1, γ_2, and γ_3, Vapor Mole Fractions y_1 and y_2, Excess Gibbs Energy G^E, Pressure P_{calc}, Boiling Temperature T_{calc}, and Differences in Pressure ΔP and Boiling Temperature ΔT

experimental data			calculated values									
liquid mole fraction		boiling temp	activity coefficient			vapor mole fraction		Gibbs energy	pressure		boiling temp	
x_1	x_2	T_{exp}/K	γ_1	γ_2	γ_3	y_1	y_2	G^E/J·mol^{-1}	P_{calc}/kPa	ΔP/kPa	T_{calc}/K	ΔT/K
						$P = 79.99$ kPa						
0.254	0.254	410.00	1.185	1.508	1.245	0.311	0.330	255.5	79.95	−0.04	410.02	0.02
0.409	0.408	408.53	1.059	1.134	1.876	0.429	0.381	189.5	80.15	0.16	408.46	−0.07
0.186	0.185	411.85	1.289	1.790	1.112	0.261	0.301	226.1	80.06	0.07	411.82	−0.03
0.563	0.281	408.13	1.036	1.174	1.864	0.572	0.269	162.2	80.05	0.06	408.11	−0.02
0.409	0.204	409.05	1.122	1.427	1.342	0.462	0.244	233.3	79.95	−0.04	409.07	0.02
0.197	0.099	413.73	1.367	2.072	1.063	0.307	0.196	176.7	80.19	0.20	413.65	−0.08
0.330	0.165	410.31	1.196	1.620	1.201	0.410	0.232	231.1	80.16	0.17	410.24	−0.07
0.484	0.344	408.66	1.047	1.153	1.858	0.499	0.325	177.7	80.91	0.92	408.27	−0.39
0.712	0.192	408.16	1.016	1.191	2.001	0.709	0.187	111.7	80.18	0.19	408.08	−0.08
0.208	0.528	409.34	1.099	1.141	1.745	0.231	0.507	236.3	80.35	0.36	409.19	−0.15
0.400	0.400	408.45	1.062	1.147	1.821	0.420	0.378	198.5	79.98	−0.01	408.46	0.01
0.571	0.286	408.21	1.034	1.164	1.909	0.579	0.272	154.9	80.20	0.21	408.12	−0.09
0.250	0.500	408.91	1.090	1.142	1.761	0.274	0.478	229.3	79.81	−0.18	408.99	0.08
0.334	0.333	409.00	1.101	1.277	1.486	0.368	0.354	245.4	80.39	0.40	408.84	−0.16
0.572	0.142	408.40	1.070	1.376	1.469	0.606	0.161	194.3	79.84	−0.15	408.46	0.06
0.287	0.142	410.78	1.248	1.755	1.144	0.382	0.222	220.1	79.20	−0.79	411.10	0.32
0.200	0.400	409.33	1.140	1.302	1.419	0.232	0.442	271.5	79.63	−0.36	409.48	0.15
0.630	0.185	408.06	1.035	1.253	1.715	0.638	0.189	163.0	80.01	0.02	408.05	−0.01
0.774	0.113	408.16	1.014	1.258	1.881	0.768	0.116	107.9	80.22	0.23	408.06	−0.10
						$P = 53.33$ kPa						
0.254	0.254	396.45	1.219	1.569	1.267	0.325	0.344	280.9	53.32	−0.01	396.45	0.00
0.409	0.408	394.76	1.068	1.148	2.006	0.435	0.382	210.6	53.40	0.07	394.72	−0.04
0.186	0.185	398.38	1.338	1.886	1.129	0.276	0.318	247.7	53.47	0.14	398.30	−0.08
0.563	0.281	394.31	1.043	1.191	2.003	0.577	0.270	181.0	53.34	0.01	394.31	−0.00
0.409	0.204	395.42	1.144	1.475	1.378	0.476	0.251	258.6	53.33	0.00	395.42	0.00
0.197	0.099	400.21	1.426	2.205	1.068	0.329	0.211	194.4	53.46	0.13	400.13	−0.08
0.330	0.165	396.69	1.230	1.693	1.219	0.428	0.242	255.3	53.45	0.12	396.62	−0.07
0.484	0.344	394.78	1.055	1.169	1.991	0.505	0.326	198.1	53.82	0.49	394.50	−0.28
0.712	0.192	394.25	1.019	1.209	2.190	0.711	0.186	125.2	53.43	0.10	394.19	−0.06
0.208	0.528	395.77	1.115	1.159	1.835	0.237	0.514	261.0	53.65	0.32	395.58	−0.19
0.400	0.400	394.71	1.072	1.163	1.939	0.427	0.380	220.5	53.29	−0.04	394.73	0.02
0.571	0.286	394.40	1.040	1.180	2.060	0.584	0.272	172.9	53.49	0.16	394.31	−0.09
0.250	0.500	395.27	1.106	1.159	1.856	0.281	0.483	253.6	53.18	−0.15	395.35	0.08
0.334	0.333	395.41	1.121	1.309	1.539	0.378	0.362	271.3	53.66	0.33	395.22	−0.19
0.572	0.142	394.68	1.084	1.417	1.529	0.618	0.164	217.2	53.23	0.10	394.74	0.06
0.287	0.142	397.23	1.289	1.845	1.156	0.401	0.234	242.7	52.84	−0.49	397.51	0.28
0.200	0.400	395.81	1.166	1.339	1.458	0.241	0.455	298.4	53.08	−0.25	395.95	0.14
0.630	0.185	394.26	1.042	1.278	1.824	0.645	0.190	182.6	53.33	0.00	394.26	0.00
0.774	0.113	394.26	1.017	1.283	2.043	0.771	0.116	121.7	53.45	0.12	394.18	−0.08
						$P = 26.66$ kPa						
0.254	0.254	375.66	1.276	1.666	1.297	0.351	0.365	319.4	26.62	−0.04	375.70	0.04
0.409	0.408	373.68	1.086	1.172	2.218	0.450	0.385	244.1	26.58	−0.08	373.76	0.08
0.186	0.185	377.61	1.420	2.040	1.141	0.304	0.348	280.2	26.71	0.05	377.57	−0.04
0.563	0.281	373.19	1.055	1.220	2.235	0.591	0.271	211.1	26.61	−0.05	373.25	0.06
0.409	0.204	374.51	1.183	1.555	1.431	0.504	0.263	297.5	26.62	−0.04	374.55	0.04
0.197	0.099	379.34	1.523	2.423	1.075	0.367	0.236	221.0	26.62	−0.04	379.38	0.04
0.330	0.165	375.83	1.288	1.811	1.245	0.461	0.259	292.0	26.72	0.06	375.77	−0.06
0.484	0.344	373.54	1.070	1.195	2.210	0.519	0.328	230.4	26.71	0.05	373.49	−0.05
0.712	0.192	372.99	1.025	1.238	2.523	0.719	0.186	147.1	26.67	0.01	372.98	−0.01
0.208	0.528	374.95	1.145	1.190	1.973	0.250	0.526	299.4	26.75	0.09	374.86	−0.09
0.400	0.400	373.70	1.091	1.189	2.126	0.443	0.384	255.1	26.55	−0.11	373.81	0.11
0.571	0.286	373.21	1.051	1.206	2.314	0.596	0.273	201.8	26.65	−0.01	373.22	0.01
0.250	0.500	374.41	1.133	1.189	2.004	0.295	0.493	291.4	26.49	−0.17	374.58	0.17
0.334	0.333	374.59	1.155	1.362	1.618	0.399	0.374	311.1	26.82	0.16	374.43	−0.16
0.572	0.142	373.63	1.108	1.484	1.621	0.642	0.170	253.0	26.54	−0.12	373.75	0.12
0.287	0.142	376.23	1.359	1.992	1.173	0.437	0.253	277.0	26.23	−0.43	376.66	0.43
0.200	0.400	375.09	1.212	1.399	1.516	0.259	0.477	339.2	26.46	−0.20	375.30	0.21
0.630	0.185	373.15	1.055	1.320	2.003	0.660	0.193	213.7	26.61	−0.05	373.20	0.05
0.774	0.113	372.98	1.022	1.324	2.323	0.780	0.117	144.1	26.67	0.01	372.98	0.00

Results and Discussion

In Table 2, the liquid-phase mole fraction x_1 and boiling temperature measurements at the pressures 26.66, 53.33, and 79.99 kPa for binary systems are presented. Table 3 presents the correlation results of $T-x$ data for the systems *o*-xylene + cyclohexanol, nonane + cyclohexanol, and earlier investigation[5] of *o*-xylene + nonane, together with coefficients of the modified Wilson equation (eqs 7 and 8). The standard deviation is expressed as

$$\sigma P = [\{\sum (P_{calc} - P_{exp})^2\}/(N - n)]^{1/2} \qquad (11)$$

where N is the number of experiments and n is the number of coefficients ($n = 4$) found.

The vapor pressures of pure components were calculated by the Antoine equation

$$\ln(P_i^\circ/\mathrm{kPa}) = A_i - \frac{B_i}{T/\mathrm{K} + C_i} \qquad (12)$$

where A_i, B_i, and C_i and the used temperature regions are reported in Table 4. For *o*-xylene and nonane the coefficients of the Antoine equation have been published earlier;[5] for the calculation of coefficients for cyclohexanol, the experimental data of this work were used. Calculated vapor pressures are in good agreement with literature data.[9]

Each binary system exhibits a minimum boiling azeotrope. The azeotropic points have been determined from the function

$$\alpha_{12} = \frac{y_1/y_2}{x_1/x_2} \qquad (13)$$

where y_i is the mole fraction of component i in the vapor phase, calculated by the modified Wilson equation and solving for $\alpha_{12} = 1$. Azeotropic compositions x^{az}, boiling temperatures T_{az}, and pressures are presented in Table 5. The azeotropic data for *o*-xylene + cyclohexanol at 101.33 kPa are given in the literature,[10] and those at 101.32, 13.33, and 1.33 kPa were reported by Garber et al.[11]

In Table 6 the isobaric VLE data for the ternary system are compared with correlation results obtained by the modified Wilson equation. We have considered ideal behavior for the vapor phase; the error in the calculation of vapor composition can be estimated to be less than 0.002. Calculations were made using the values of coefficients for three constituent binaries given in Table 3. The mean absolute relative error for all three pressures was equal to 0.32%. Calculation of VLE for the ternary system in the boiling temperature region (373 to 414 K) leads to nearly perfect agreement with experimental data, as seen from differences between experimental and calculated values of pressure and temperature.

The ternary system exhibits a minimum boiling azeotrope whose composition at three pressures was determined by the minimization of the function

$$(\alpha_{ij} - 1) + (\alpha_{ik} - 1) \Rightarrow \min \qquad (14)$$

The parameters of the ternary azeotrope *o*-xylene + nonane + cyclohexanol obtained at three pressures are given in Table 7.

Table 7. Azeotropic Boiling Temperatures T_{az} and Compositions x_i^{az} of the Ternary System *o*-Xylene (1) + Nonane (2) + Cyclohexanol (3) versus Pressure *P*

P/kPa	T_{az}/K	x_1^{az}	x_2^{az}
26.66	372.94	0.765	0.142
53.33	394.15	0.715	0.163
79.99	408.01	0.685	0.175

As seen from data obtained for the ternary system, the modified Wilson equation gives a good correlation in a large boiling temperature range, simultaneously taking into account all experiments carried out at different pressures. Evidently, a better agreement will be achieved also between VLE and excess enthalpy data, usually obtained at different temperatures.[12]

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