Vapor−Liquid Equilibrium of Binary and Ternary Mixtures Containing Isopropyl Ether, 2-Butanol, and Benzene at T = 313.15 K

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Experimental isothermal $P-x$ data for the ternary system isopropyl ether + 2-butanol + benzene and for the binary systems isopropyl ether + 2-butanol and 2-butanol + benzene at T = 313.15 K are reported. A static technique consisting of an isothermal total pressure cell was used for the measurements. Data reduction by Barker's method provides correlations for G^E, using the Margules equation for the binary systems and the Wohl expansion for the ternary system. Wilson, NRTL, and UNIQUAC models have been applied successfully to binary and ternary systems. The mixtures measured exhibit positive deviation from ideal behavior. The binary system benzene + 2-butanol exhibits positive azeotropy, and no ternary azeotrope has been detected in the ternary mixture.

Introduction

The formulation of new gasolines, according to the environmental restrictions, means the use of ethers and alcohols as blending agents for enhancing the octane number and a modification of the refinery. Simulation of the processes is the first step in the design or optimization of any stage of a plant. The accuracy of a process simulation depends strongly on the thermodynamic models used to describe the physical behavior of the involved components. The highest quality of vapor−liquid equilibria data are required to improve the parameters of the predictive models that are used in process simulation packages.

Our group has undertaken a research program on the thermodynamic characterization of ternary mixtures as the simplest multicomponent system, containing oxygenated additives (ethers and alcohols) and different types of hydrocarbons (paraffins, cycloparaffins, aromatics, oleffins), to better understand and model these reformulated gasolines. Methyl *tert*-butyl ether (MTBE), *tert*-amylmethyl ether (TAME), and isopropyl ether or diisopropyl ether (DIPE) were chosen as representative ethers; methanol, 2-propanol, 1-propanol, and *tert*-amyl alcohol were chosen as alcohol additives.

We have now started a series of measurements of ternary mixtures containing DIPE + benzene + butanol isomers; the results of mixtures containing 1-butanol[1] and 2-butanol[2] are in press. In this work, experimental isothermal $P-x$ data are reported for the ternary system isopropyl ether (DIPE) + 2-butanol + benzene and for two of the binary systems DIPE + 2-butanol and benzene + 2-butanol at 313.15 K.

Experimental Section

Materials. Isopropyl ether and benzene were purchased from Fluka Chemie AG and were of the highest purity available, chromatography quality reagents (of the series puriss. p.a.) with a purity > 0.99 and > 0.995 (by gas chromatography, GC), respectively. 2-Butanol used was anhydrous Aldrich product with a purity > 0.999 (GC). All liquids were thoroughly degassed before measurements by a modified distillation method based on the one suggested by Van Ness and Abbott[3] and kept

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Table 1. Average Values of Experimental Vapor Pressures (p_i^{sat}) for the Pure Compounds Measured in This Work and Literature Values (p_i^{sat}(lit)), Molar Volumes of Pure Liquids (V_i^L), Second Virial Coefficients (B_{ii}, B_{ij}), and van der Waals Molecular Volumes (r_i) and Surfaces (q_i) at T = 313.15 K Used for the Calculations

	isopropyl ether (i = 1)	2-butanol (i = 2)	benzene (i = 3)
p_i^{sat}/(kPa)	37.108	6.055	24.386
p_i^{sat}(lit)/(kPa)	37.128[a]	6.048[d]	24.398[a,g]
	37.090[b]	6.046[e]	24.367[h]
	37.081[c]	6.017[f]	24.380[i]
V_i^L/(cm³·mol⁻¹)[j]	145	94	91
B_{i1}/(cm³·mol⁻¹)[k]	−1687.8	−1053.6	−1701.0
B_{i2}/(cm³·mol⁻¹)[k]	−1608.0	−3241.8	−1053.6
B_{i3}/(cm³·mol⁻¹)[k]	−1701.0	−1053.6	−1310.5
r_i[l]	4.7421	3.9235	3.1878
q_i[l]	4.088	3.664	2.400

a Ref 18. *b* Ref 19. *c* Calculated from the Antoine equation using constants reported in ref 20. *d* Ref 21. *e* Ref 22. *f* Ref 23. *g* Ref 24. *h* Ref 25. *i* Ref 26. *j* Ref 27. *k* Calculated by Hayden et al.[11] from Dymond et al.[12] *l* Calculated from ref 28.

in glass balloons equipped with leak-proof valves. The purities of the chemicals were checked by GC and were found to be > 0.995 for all the compounds. In Table 1, the vapor pressures of the pure constituents measured in this work are compared with those reported in the literature as a check for complete degassing.

Apparatus and Procedure. A static VLE apparatus, consisting of an isothermal total pressure cell, has been employed for measuring the vapor−liquid equilibrium of binary and ternary mixtures. The apparatus and measuring technique are based on that by Van Ness and co-workers[4,5] and whose performance has been described in a previous paper.[6,7]

Experimental values of total vapor pressure for the binary mixtures were obtained in two overlapping runs starting from opposite ends of the composition range. For the ternary mixture, data were obtained by the addition of a pure species to a mixture of the other two at a fixed temperature. Six runs (dilution lines) were made starting from the corresponding binary system at mole fractions close to 0.3 or 0.7 and adding the third pure component up to a mole fraction of 0.5.

The equilibrium properties measured directly and their uncertainties are as follows: injected volume ± 0.03 mL,

temperature $\pm$ 10 mK, and total pressure $\pm$ 5 Pa. The overall mole fraction is determined accurately from the volumetric displacement of the pistons, the temperature of the injectors, and the densities for the pure components. This resulted in uncertainties in mole fraction of $\pm$ 0.0005, the composition of the liquid phase is close to the overall composition, but it is corrected taken into account the vapor space of cell and using the virial equation.

Experimental Results and Correlations

Data reduction for the binary and ternary mixtures was done by Barker's method[8] according to well-established procedures.[9,10]

The nonideality of the vapor phase was taken into account with the virial equation of state, truncated after the second term. The pure component and interaction second virial coefficients (B_{ij}) were calculated by the Hayden and O'Connell method[11] using the parameters given by Dymond and Smith.[12] They appear in Table 1.

Binary systems have been correlated by a five-parameter Margules[13] equation:

$$g_{ij} = \frac{G_m^E}{RT} = \{A_{ji}x_i + A_{ij}x_j - (\lambda_{ji}x_i + \lambda_{ij}x_j)x_ix_j + \eta x_i^2 x_j^2\}x_ix_j \quad (1)$$

where parameters A_{ij}, A_{ji}, λ_{ij}, λ_{ji}, and η were obtained by regression of the binary data. The ternary system has been correlated by three-parameter Wohl equation,[14] which also includes the parameters of the corresponding binary systems (g_{ij}) previously obtained by eq 1:

$$g_{123} = \frac{G^E}{RT} = g_{12} + g_{13} + g_{23} + (C_0 + C_1x_1 + C_2x_2)x_1x_2x_3 \quad (2)$$

where only parameters C_0, C_1, and C_2 were found by regression of the ternary data.

Also the Wilson,[15] NRTL,[16] and UNIQUAC[17] models have been used for fitting both binary and ternary systems, whose expressions for the excess Gibbs energy are given by

$$\frac{G^E}{RT} = -\sum_i x_i \ln(\sum_j x_j A_{ij}) \quad (3)$$

$$\frac{G^E}{RT} = \sum_i x_i \frac{\sum_j A_{ji}G_{ji}x_j}{\sum_k G_{ki}x_k} \quad (4)$$

$$\frac{G^E}{RT} = \sum_i x_i \ln\frac{\varphi_i}{x_i} + \frac{z}{2}\sum_i q_ix_i \ln\frac{\vartheta_i}{q_i} - \sum_i q_ix_i \ln(\sum_j \vartheta_j A_{ji}) \quad (5)$$

where $G_{ji} = \exp(-\alpha_{ji}A_{ji})$, $\vartheta_i = q_ix_i/\sum_j q_jx_j$, $\varphi_i = r_ix_i/\sum_j r_jx_j$, and $z = 10$; the adjustable parameters are A_{ij} and A_{ji}. The parameter α_{ji} of the NRTL model has also been adjusted.

Tables 2 and 3 show experimental values of total pressure and the calculated mole fractions of the liquid and vapor for the binary systems and the ternary system. In these tables, the compositions of the vapor phases were calculated by the Margules equation for the binary systems and the Wohl expansion for the ternary system.

Results of data correlation for the binary systems are summarized in Table 4. It contains the results of the correlation of the other binary system that have been published previously.[18]

Table 2. Total Pressure (p) for the Binary Systems at T = 313.15 K and at Various Compositions of the Liquid Phase (x_1) and the Calculated Composition of the Vapor Phase (y_1) Using a Five-Parameter Margules Equation

x_1	y_1	p/(kPa)	x_1	y_1	p/(kPa)
Isopropyl Ether (1) + 2-Butanol (2)					
0.0000	0.0000	6.050	0.5009	0.8652	28.235
0.0558	0.4424	10.314	0.5495	0.8767	29.270
0.0999	0.5831	13.253	0.5529	0.8775	29.345
0.1537	0.6782	16.337	0.5925	0.8862	30.128
0.2025	0.7316	18.746	0.6030	0.8884	30.316
0.2508	0.7684	20.807	0.6515	0.8985	31.199
0.3007	0.7967	22.675	0.7006	0.9086	32.045
0.3516	0.8192	24.342	0.7506	0.9191	32.885
0.4000	0.8366	25.734	0.8005	0.9303	33.702
0.4010	0.8369	25.774	0.8519	0.9434	34.547
0.4504	0.8519	27.052	0.9003	0.9580	35.356
0.4507	0.8520	27.055	0.9531	0.9777	36.275
0.4998	0.8649	28.212	1.0000	1.0000	37.103
Benzene (1) + 2-Butanol (2)					
0.0000	0.0000	6.057	0.5011	0.8180	22.119
0.0594	0.4274	10.024	0.5486	0.8292	22.630
0.1087	0.5630	12.574	0.5507	0.8297	22.655
0.1457	0.6243	14.184	0.5989	0.8403	23.111
0.1958	0.6810	16.027	0.6015	0.8408	23.134
0.2552	0.7261	17.787	0.6513	0.8514	23.545
0.2978	0.7498	18.826	0.7028	0.8622	23.919
0.3486	0.7721	19.861	0.7554	0.8734	24.258
0.4006	0.7903	20.768	0.8020	0.8839	24.507
0.4022	0.7909	20.782	0.8522	0.8970	24.707
0.4476	0.8042	21.446	0.8994	0.9134	24.855
0.4507	0.8051	21.492	0.9521	0.9435	24.837
0.4997	0.8177	22.101	1.0000	1.0000	24.398

For the ternary system, the results of the correlation are given in Table 5. Both tables contain the adjustable parameter values of the different models that lead to the results using Barker's method, the root-mean-square of the difference between the experimental and the calculated pressures (rmsd Δp), and the maximum value of this difference (max $|\Delta p|$).

Figure 1 shows a plot of ($p_{exp} - p_{calc}$) against x_1 for the binary systems, where the pressures were calculated by the Margules equation. It can be seen that both branches, necessary to cover the entire composition range, exhibit good agreement close to equimolar concentrations. Furthermore, all deviations are less than 0.1 % of the total pressure.
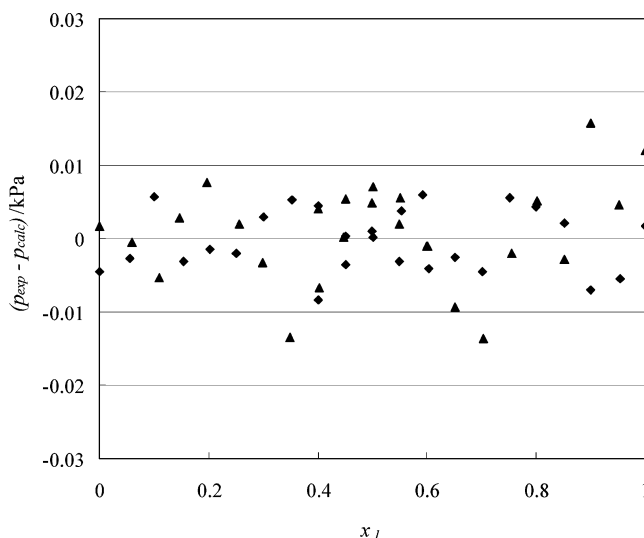

Figure 1. Pressure residuals ($p_{exp} - p_{calc}$) defined as differences between experimental and calculated pressures as a function of the liquid composition (x_1): ◆, diisopropyl ether (1) + 2-butanol (2); ▲, benzene (1) + 2-butanol (2).

Table 3. Total Pressure (p) for the Ternary System Isopropyl Ether (1) + 2-Butanol (2) + Benzene (3) at $T = 313.15$ K and at Various Compositions of the Liquid (x_1, x_2) and the Vapor Phases (y_1, y_2) Calculated Using Wohl Expansion

x_1	x_2	y_1	y_2	p/kPa	x_1	x_2	y_1	y_2	p/kPa
1.0000	0.0000	1.0000	0.0000	37.141	1.0000	0.0000	1.0000	0.0000	37.120
0.7018	0.2982	0.9088	0.0912	32.125	0.7020	0.0000	0.7670	0.0000	34.207
0.6853	0.2912	0.8858	0.0904	32.015	0.6843	0.0252	0.7537	0.0150	33.856
0.6662	0.2831	0.8594	0.0894	31.892	0.6673	0.0494	0.7427	0.0272	33.510
0.6317	0.2684	0.8121	0.0877	31.665	0.6315	0.1005	0.7240	0.0480	32.795
0.5969	0.2536	0.7653	0.0861	31.422	0.5972	0.1492	0.7101	0.0634	32.120
0.5623	0.2388	0.7198	0.0845	31.176	0.5680	0.1909	0.7001	0.0744	31.547
0.5273	0.2239	0.6747	0.0829	30.917	0.5272	0.2489	0.6881	0.0878	30.730
0.4922	0.2090	0.6304	0.0812	30.657	0.4915	0.2999	0.6787	0.0986	29.984
0.4562	0.1937	0.5858	0.0795	30.375	0.4554	0.3513	0.6695	0.1092	29.196
0.4209	0.1787	0.5429	0.0776	30.087	0.4213	0.3999	0.6609	0.1195	28.393
0.3862	0.1640	0.5013	0.0757	29.791	0.3861	0.4500	0.6515	0.1308	27.507
0.3509	0.1489	0.4594	0.0735	29.480	0.3512	0.4997	0.6414	0.1431	26.550
0.0000	1.0000	0.0000	1.0000	6.062	0.0000	1.0000	0.0000	1.0000	6.063
0.3025	0.6975	0.7976	0.2024	22.728	0.0000	0.6966	0.0000	0.2474	18.951
0.2921	0.6734	0.7357	0.1952	23.107	0.0322	0.6742	0.0862	0.2321	19.821
0.2871	0.6620	0.7081	0.1920	23.276	0.0595	0.6551	0.1534	0.2199	20.528
0.2703	0.6231	0.6219	0.1813	23.791	0.1024	0.6252	0.2480	0.2024	21.620
0.2574	0.5931	0.5633	0.1737	24.140	0.1566	0.5875	0.3501	0.1831	22.964
0.2424	0.5585	0.5029	0.1657	24.487	0.2005	0.5568	0.4207	0.1695	23.993
0.2271	0.5231	0.4484	0.1584	24.788	0.2506	0.5219	0.4898	0.1559	25.093
0.2118	0.4879	0.4001	0.1517	25.043	0.3035	0.4850	0.5524	0.1433	26.185
0.1969	0.4534	0.3578	0.1458	25.253	0.3499	0.4527	0.6000	0.1335	27.086
0.1815	0.4180	0.3187	0.1401	25.427	0.4000	0.4177	0.6455	0.1239	28.018
0.1667	0.3837	0.2842	0.1349	25.556	0.4498	0.3831	0.6856	0.1151	28.873
0.1527	0.3516	0.2544	0.1301	25.657	0.5001	0.3481	0.7221	0.1067	29.710
0.0000	0.0000	0.0000	0.0000	24.392	0.0000	0.0000	0.0000	0.0000	24.379
0.3005	0.0000	0.4060	0.0000	29.503	0.0000	0.2991	0.0000	0.1382	23.894
0.2931	0.0247	0.3922	0.0211	29.396	0.0348	0.2887	0.0593	0.1312	24.489
0.2857	0.0491	0.3812	0.0375	29.219	0.0515	0.2837	0.0864	0.1281	24.771
0.2705	0.0998	0.3641	0.0622	28.792	0.1003	0.2691	0.1616	0.1195	25.573
0.2556	0.1495	0.3518	0.0793	28.342	0.1517	0.2537	0.2345	0.1113	26.376
0.2407	0.1992	0.3420	0.0927	27.873	0.1994	0.2394	0.2971	0.1044	27.091
0.2254	0.2501	0.3334	0.1045	27.357	0.2499	0.2243	0.3587	0.0975	27.821
0.2103	0.3004	0.3257	0.1153	26.810	0.2996	0.2094	0.4156	0.0911	28.517
0.1954	0.3500	0.3182	0.1258	26.228	0.3495	0.1945	0.4693	0.0849	29.189
0.1805	0.3995	0.3108	0.1366	25.603	0.3993	0.1796	0.5199	0.0788	29.841
0.1652	0.4502	0.3029	0.1483	24.898	0.4498	0.1645	0.5687	0.0728	30.493
0.1513	0.4965	0.2954	0.1600	24.193	0.4999	0.1495	0.6148	0.0669	31.123

Discussion

We have not found literature data available for comparison for any of the binary and ternary systems at $T = 313.15$ K presented in this paper.
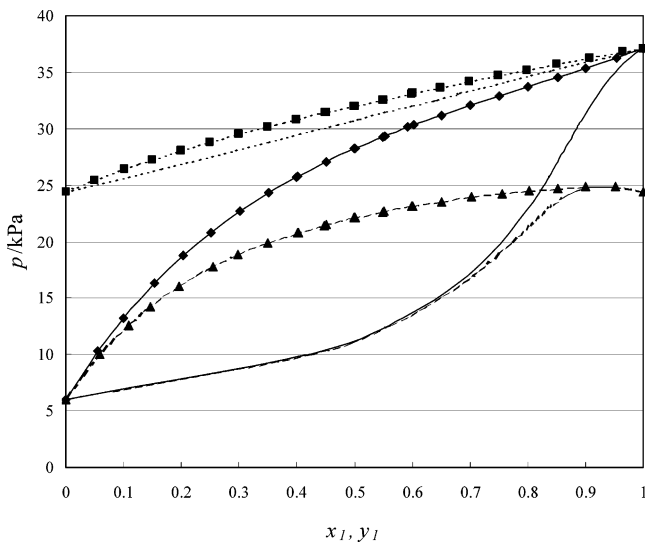
The two binary systems measured exhibit a positive deviation from ideality. This effect is stronger for benzene + 2-butanol, which presents an azeotrope. Figure 2 is an $p-x-y$ plot where the three binary subsystems involved in the ternary mixture are shown.

The five-parameter Margules equation leads to the best correlation results for the two binary systems measured in this
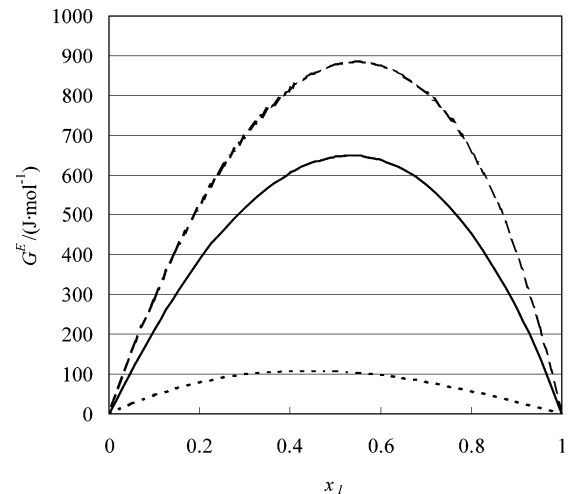

Figure 2. Total pressure at $T = 313.15$ K of the binary systems as a function of the liquid (x_1) and vapor composition (y_1): ◆, isopropyl ether (1) + 2-butanol (2); ▲, benzene (1) + 2-butanol (2); and ■, isopropyl ether (1) + benzene (2). Symbols represent the experimental points; lines are the calculations of Margules equation.

Figure 3. Excess Gibbs energy, calculated by Margules equation, for the three binary systems as a function of the liquid mole fraction (x_1): —, isopropyl ether (1) + 2-butanol (2); - - -, benzene (1) + 2-butanol (2); and ⋯, isopropyl ether (1) + benzene (2).

Table 4. Parameters of the Models Used for the Binary Subsystems of Ternary System Isopropyl Ether (1) + 2-Butanol (2) + Benzene (3) at $T = 313.15$ K, Together with the Root Mean Square Deviation of Pressure (rmsd Δp) and the Maximum Value of the Deviation (max $|\Delta p|$)[a]

	Margules	Wilson	NRTL	UNIQUAC
Isopropyl Ether (1) + 2-Butanol (2)				
A_{12}	0.8803	0.7503	0.8812	0.5728
A_{21}	1.1796	0.3911	0.3891	1.2528
λ_{12}	0.0096			
λ_{21}	0.1588			
η	−0.2729			
α_{12}			0.6094	
rmsd Δp/kPa	0.005	0.016	0.013	0.055
max $|\Delta p|$/kPa	0.008	0.030	0.040	0.088
Benzene (1) + 2-Butanol (2)				
A_{12}	1.2439	0.6736	1.4611	0.7956
A_{21}	1.9499	0.2190	0.6823	0.8655
λ_{12}	0.8577			
λ_{21}	1.8142			
η	1.3643			
α_{12}			0.6506	
rmsd Δp/kPa	0.007	0.087	0.037	0.190
max $|\Delta p|$/kPa	0.020	0.145	0.062	0.301
x_1, azeotrope	0.9289	0.9310	0.9273	0.9373
p azeotrope	24.859	24.755	24.822	24.624
Isopropyl Ether (1) + Benzene (2)[b]				
A_{12}	0.2134	0.5515	−0.4967	0.9014
A_{21}	0.1277	1.3762	0.7905	1.0698
$\lambda_{12} = \lambda_{21}$	0.0282			
α_{12}			0.3	
rmsd Δp/kPa	0.005	0.006	0.005	0.006
max $|\Delta p|$/kPa	0.009	0.013	0.008	0.013

[a] The Δp term is defined as the difference between the experimental and calculated pressure. [b] Data published in ref 18.

Table 5. Parameters of the Models Used for the Ternary System Isopropyl Ether (1) + 2-Butanol (2) + Benzene (3) at $T = 313.15$ K, Together with the Root Mean Square Deviation of Pressure (rmsd Δp) and the Maximum Value of the Deviation (max $|\Delta p|$)[a]

	Wilson	NRTL	UNIQUAC	Wohl
A_{12}	0.8005	0.9075	0.4422	$C_0 = 2.5880$
A_{21}	0.3593	0.3730	1.4121	$C_1 = 0.3372$
A_{13}	0.3739	−0.5658	1.2008	$C_2 = -0.9589$
A_{31}	1.6776	0.9030	0.7800	
A_{23}	0.2181	0.7100	1.1299	
A_{32}	0.6819	1.4302	0.5082	
α_{12}		0.6094		
α_{13}		0.3000		
α_{23}		0.6506		
rmsd Δp/kPa	0.033	0.013	0.049	0.032
max $|\Delta p|$/kPa	0.094	0.033	0.171	0.058

[a] The Δp term is defined as the difference between the experimental and calculated pressure.

work. The root-mean-square deviation of the pressure is 5 Pa for (isopropyl ether + 2-butanol) and 7 Pa for (benzene + 2-butanol), and the maximum deviations are 8 Pa and 20 Pa, respectively. The other models give higher values of the root-mean-square deviations. The system containing benzene presents a maximum pressure azeotrope. Its corresponding composition and pressure have been calculated for all the models, and these values are summarized in Table 4. The Margules equation predicts the azeotrope for a mole fraction of benzene of 0.9289 and a pressure of 24.859 kPa.

Finally, we have calculated the excess molar Gibbs energy for the binary systems. Figure 3 shows the values of G_m^E, calculated by the Margules equation, as a function of the liquid mole fraction. Again, it may be noticed that the system {benzene (1) + 2-butanol (2)} gives the maximum value of 885 J·mol^{-1} for G_m^E at a composition around $x_1 = 0.55$. On the other hand,
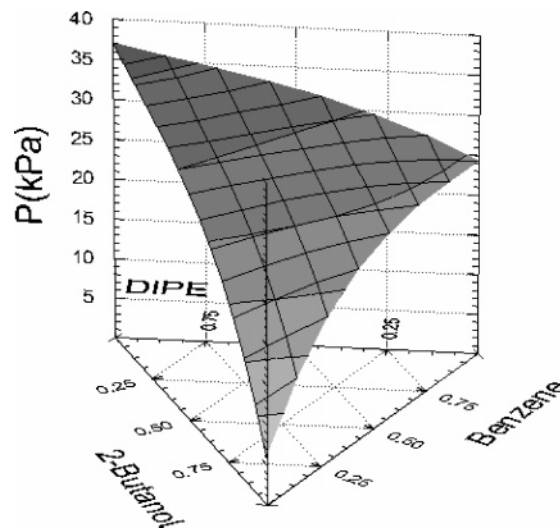

Figure 4. Oblique view of the pressure surface reduced by Wohl expansion for the ternary system isopropyl ether (DIPE) (1) + 2-butanol (2) + benzene (3) at $T = 313.15$ K.
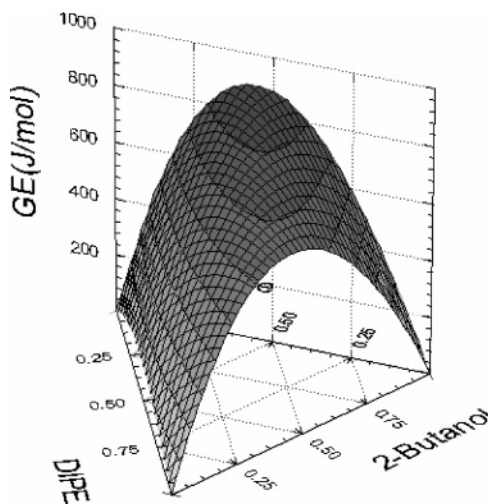

Figure 5. Oblique view of the excess Gibbs energy surface reduced by Wohl expansion for the ternary system isopropyl ether (DIPE) (1) + 2-butanol (2) + benzene (3) at $T = 313.15$ K.

the system {isopropyl ether (1) + 2-butanol (2)} presents a maximum of 648 J·mol^{-1} for G_m^E at a composition around $x_1 = 0.55$.

The measurements for the system isopropyl ether (DIPE) + 2-butanol + benzene are well-correlated by all the models. The root-mean-square pressure deviation varies from 13 Pa using the NRTL model to 52 Pa for UNIQUAC model, with a maximum value of the absolute deviation in pressure of 33 Pa for the NRTL model up to 147 Pa for UNIQUAC, within a maximum range of pressure close to 37000 Pa. Also good results are obtained in the prediction of the total pressure for the ternary system using the parameters of the binary systems summarized in Table 4 for Wilson, NRTL, and UNIQUAC. The root-mean-square pressure deviations are 115 Pa, 30 Pa, and 176 Pa, respectively.

Graphical results for the ternary system are in Figures 4 and 5. They show the oblique view of the pressure and of the excess Gibbs energy surface respectively, calculated by Wohl expansion.

The pressure surface increases from the value of the saturation pressure of a less volatile compound (2-butanol) to the saturation pressure of a more volatile compound (isopropyl ether). Around

the binary azeotrope, the surface shows a slight plateau. The ternary system also shows a positive deviation from ideality. The molar excess Gibbs energy increases up to a maximum value, which corresponds to a less ideal binary system.

Literature Cited

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