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Vapor−Liquid Equilibria of Binary Mixtures Containing 1-Butanol and Hydrocarbons at 313.15 K

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ABSTRACT: Total pressure measurements made at 313.15 K by the static method for five binary mixtures 1-butanol + heptane, 2,2,4-trimethylpentane, dodecane, cyclohexane, and methylbenzene are presented. Data reduction by Barker's method provides correlations for G^E, using the four-parameter Margules equation; also the Wilson, nonrandom two-liquid (NRTL), and universal quasichemical activity coefficient (UNIQUAC) models have been used for fitting binary systems. The five investigated mixtures exhibit a positive deviation from ideality. No azeotrope has been detected for 1-butanol + dodecane, and the other systems show positive azeotropy. Good results are obtained in the prediction of total pressure for these systems.

■ INTRODUCTION

The formulation of new gasolines, according to environmental restrictions, requires the use of ethers and alcohols as blending agents for enhancing the octane number and a modification of the refinery. The simulation of these processes is the first step in the design or optimization of any stage of a production 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 (VLE) data are required to improve the parameters of the predictive models that are used in process simulation packages.

The present work is part of a research program on the VLE of binary and ternary mixtures containing 1-butanol or 2-butanol with several hydrocarbons.

In this paper, the experimental isothermal ($P-x$) data are reported for the binary systems 1-butanol + heptane, 2,2,4-trimethylpentane, dodecane, cyclohexane, and methylbenzene at 313.15 K.

■ EXPERIMENTAL SECTION

Materials. 1-Butanol, heptane, and cyclohexane were supplied from Fluka Chemie and were of highest purity available, with a purity (in mole fraction) of > 0.998, > 0.998, and > 0.999 (by gas chromatography, GC), respectively. Methylbenzene was an anhydrous Aldrich product with a purity of >0.999, for the other compounds (2,2,4-trimethylpentane and dodecane) were supplied from Sigma-Aldrich with a purity of >0.997. All liquids were thoroughly degassed before measurements by a modified distillation method based on the one suggested by Van Ness and Abbott[1] and kept in glass balloons equipped with leak-proof valves. 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 VLE of binary and ternary mixtures. The apparatus and measuring technique were based on those of Van Ness and co-workers.[2,3]

The sample injectors were three 100 cm^3 positive displacement pumps (Ruska, model 2200-801) with a resolution of 0.01 cm^3 and an estimated total uncertainty of ± 0.03 cm^3. These allowed the injection of known volumes of the pure components, previously degassed, into the cell which was immersed in a high precision water bath (Hart Scientific model 6020), assuring a stability of ± 0.5 mK when thermostatted at T = 313.15 K.

The cell was a cylindrical stainless steel piece with a capacity of about 180 cm^3 fitted with a magnetic stirrer coupled to an external drive. An initial volume of about 50 cm^3 of one component was injected into the evacuated cell, and the vapor pressure was measured. Successive injections of a second component were made over a desired composition range until the cell was nearly full. The total mass injected was determined accurately from the volumetric displacement of the pistons, the temperature of the injectors, and the known densities for the pure components. This resulted in uncertainties in mole fraction of ± 0.0001.

Experimental values of total vapor pressure for the binary mixtures were obtained in two overlapping runs starting from opposite ends of the composition range.

The temperature was measured by means of a calibrated platinum resistance thermometer (SDL model 5385/100) and an alternating current (AC) resistance bridge (ASL model F250)

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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}), van der Waals Molecular Volumes (r_i), Surfaces (q_i), and Second Virial Coefficients (B_{ii}, B_{ij}) Calculated by the Method of Hayden and O'Connell[7] at T = 313.15 K Used for the Calculations

compounds	1-butanol (i = 1)	heptane (i = 2)	2,2,4-trimethylpentane (i = 3)	dodecane (i = 4)	cyclohexane (i = 5)	methylbenzene (i = 6)
P_i^{sat}(exp)/kPa	2.502	12.326	12.986	0.059	24.629	7.877
P_i^{sat}(lit)/kPa	2.516[a]	12.323[h]	12.981[l]	0.059[s]	24.630[p]	7.875[x]
	2.464[b]	12.355[i]	12.973[p]	0.053[q]	24.632[k]	7.897[p]
	2.550[c]	12.331[j]	12.959[q]	0.054[k]	24.630[m]	7.893[a]
	2.367[d]	12.335[k]	12.960[r]		24.625[t]	7.887[k]
	2.391[e]	12.300[l]	12.966[k]		24.635[u]	7.886[q]
	2.308[f]	12.348[m]			24.652[h]	
	2.496[g]	12.334[n]			24.626[v]	
		12.338[o]			24.639[w]	
(V_i^{L}/cm$^3\cdot$mol^{-1})[y]	93	150	169	232	111	109
B_{i1}/cm$^3\cdot$mol^{-1}	−5179.5	−1451.5	−1508.4	−2940.2	−1163.7	−1336.5
B_{i2}/cm$^3\cdot$mol^{-1}	−1451.5	−2521.3				
B_{i3}/cm$^3\cdot$mol^{-1}	−1508.4		−2716.0			
B_{i4}/cm$^3\cdot$mol^{-1}	−2940.2			−16442.0		
B_{i5}/cm$^3\cdot$mol^{-1}	−1163.7				−1554.6	
B_{i6}/cm$^3\cdot$mol^{-1}	−1336.5					−2105.7
r_i	3.45[z]	5.17[z]	5.85[z]	8.55[aa]	3.97[z]	3.92[z]
q_i	3.05[z]	4.4[z]	4.94[z]	7.09[aa]	3.01[z]	2.97[z]

[a] Reported by Oracz and Kolasinska.[13] [b] Calculated according to Brown and Smith.[14] [c] Calculated according to Geiseler et al.[15] [d] Calculated from the Antoine equation using constants reported in TRC.[16] [e] Calculated from the Antoine equation using constants reported by Hiaki et al.[17] [f] Calculated from the Antoine equation using constants reported by Hauschild et al.[18] [g] Calculated from the Antoine equation using constants reported by Kemme et al.[19] [h] Calculated from the Antoine equation using constants reported by Negadi et al.[20] [i] Reported by Chamorro et al.[21] [j] Reported by Segovia et al.[22] [k] Calculated from the Antoine equation using constants reported in TRC.[23] [l] Calculated from the Antoine equation using constants reported by Reid et al.[24] [m] Reported by Montero.[25] [n] Reported by Goral.[26] [o] Reported by Lozano et al.[27] [p] Reported by Chamorro et al.[28] [q] Calculated from the Antoine equation using constants reported in NIST (coefficients calculated by NIST from ref 29). [r] Reported by Goral and Asmanova.[30] [s] Adjustment of experimental data[31] using the Antoine equation. [t] Reported by Oracz et al.[32] [u] Reported by Segovia et al.[33] [v] Calculated from the Antoine equation using constants reported by Del Río et al.[34] [w] Reported by Villamañan et al.[35] [x] Reported by Vilamañan et al.[36] [y] Reported in Riddick et al.[37] [z] Reported in the book of computer calculations for multicomponent vapor−liquid and liquid−liquid equilibria.[38] [aa] Reported by Maia de Oliveira et al.[39]

with a resolution of 1 mK and an estimated overall uncertainty of ± 10 mK. The measurement of pressure was done indirectly through a differential pressure cell and indicator (Ruska models 2413-705 and 2416-711, respectively). Air was used on the reference side of the differential pressure cell and the pressure required to obtain a null indication was measured with a fused quartz Bourdon pressure gauge (Texas Instruments model 801). The overall uncertainty of the pressure was estimated to be ± 5 Pa.

RESULTS AND CORRELATIONS

Data reduction for the binary was done by Barker's method[4] according to well-established procedures.[5,6]

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[7] using the parameters given by Dymond and Smith.[8]

The second virial coefficients (B_{ii}, B_{ij}) are reported in Table 1 together with the molar volumes of pure liquids (V_i^{L}), van der Waals molecular volumes (r_i), and surfaces (q_i).

Binary systems have been correlated by the four-parameter Margules equation:[9]

$$g_{ij} = \frac{G^{\mathrm{E}}}{x_i x_j RT} = [A_{ji}x_i + A_{ij}x_j - (\lambda_{ji}x_i + \lambda_{ij}x_j)x_i x_j] \qquad (1)$$

where parameters A_{ij}, A_{ji}, λ_{ij}, and λ_{ji} were obtained by regression of the binary data. Also the Wilson,[10] NRTL,[11] and UNIQUAC[12] models have been used for fitting binary systems, whose expressions for the excess Gibbs energy are given by

$$\frac{G^{\mathrm{E}}}{RT} = -\sum_i x_i \ln\Big(\sum_j x_j A_{ij}\Big) \qquad (2)$$

$$\frac{G^{\mathrm{E}}}{RT} = \sum_i x_i \frac{\sum_j A_{ji} G_{ji} x_j}{\sum_k G_{ki} x_k} \qquad (3)$$

$$\frac{G^{\mathrm{E}}}{RT} = \sum_i x_i \ln \frac{\varphi_i}{x_i} + \frac{z}{2} \sum_i q_i x_i \ln \frac{\vartheta_i}{q_i} - \sum_i q_i x_i \ln\Big(\sum_i \vartheta_j A_{ji}\Big) \qquad (4)$$

where $G_{ji} = \exp(-\alpha_{ji}A_{ji})$, $\vartheta_i = q_i x_i / \Sigma_j q_j x_j$, $\varphi_i = r_i x_i / \Sigma_j r_j x_j$ and z = 10. The adjustable parameters are A_{ij} and A_{ji}. The parameter α_{ji} of the NRTL model has also been adjusted.

Table 2 gives experimental values of total pressure and the corresponding compositions of the liquid and vapor phases for the five binary systems; the vapor-phase composition was calculated using the Margules equation.

Results of data correlation for all the binary systems are summarized in Table 3. This table contains the adjustable parameter

Table 2. Experimental Total Pressure (P/kPa) for the Binary Systems at T = 313.15 K at Various Compositions of the Liquid Phase (x_1) and the Calculated Compositions of the Vapor Phase (y_1) Using the Four-Parameter Margules Equation

x_1	y_1	P	x_1	y_1	P
1-Butanol (1) + Heptane (2)					
0.0000	0.0000	12.326	0.4964	0.1483	12.459
0.0518	0.0890	13.155	0.5436	0.1544	12.258
0.0958	0.1114	13.222	0.5504	0.1554	12.253
0.1459	0.1201	13.200	0.5934	0.1615	12.019
0.1945	0.1233	13.150	0.6003	0.1626	11.995
0.2437	0.1255	13.089	0.6507	0.1709	11.659
0.2939	0.1283	12.988	0.7004	0.1810	11.228
0.3433	0.1319	12.883	0.7509	0.1949	10.651
0.3933	0.1365	12.760	0.8011	0.2154	09.870
0.4000	0.1372	12.755	0.8513	0.2493	08.806
0.4432	0.1419	12.620	0.9017	0.3129	07.337
0.4501	0.1427	12.613	0.9519	0.4604	05.284
0.4932	0.1479	12.451	1.0000	1.0000	02.514
1-Butanol (1) + 2,2,4-Trimethylpentane (2)					
0.0000	0.0000	13.009	0.5012	0.1447	12.830
0.0509	0.0779	13.662	0.5504	0.1514	12.538
0.0981	0.1020	13.709	0.5524	0.1517	12.581
0.1495	0.1121	13.662	0.6002	0.1589	12.283
0.1993	0.1166	13.624	0.6339	0.1646	12.035
0.2497	0.1199	13.522	0.6508	0.1677	11.934
0.2992	0.1234	13.423	0.7005	0.1780	11.474
0.3499	0.1276	13.298	0.7511	0.1919	10.866
0.4003	0.1326	13.147	0.7999	0.2112	10.105
0.4007	0.1326	13.170	0.8493	0.2425	9.092
0.4501	0.1382	12.965	0.8969	0.2966	7.735
0.4510	0.1384	13.019	0.9528	0.4499	5.389
0.5008	0.1446	12.756	1.0000	1.0000	2.547
1-Butanol (1) + Dodecane (2)					
0.0000	0.0000	0.058	0.4995	0.9763	2.097
0.1125	0.9656	1.612	0.5298	0.9767	2.116
0.1644	0.9694	1.757	0.5504	0.9770	2.125
0.2005	0.9708	1.822	0.5803	0.9774	2.141
0.2500	0.9720	1.893	0.5998	0.9776	2.152
0.2837	0.9727	1.934	0.6310	0.9780	2.165
0.3162	0.9733	1.965	0.6828	0.9787	2.194
0.3718	0.9743	2.015	0.7346	0.9795	2.223
0.3806	0.9744	2.028	0.7861	0.9806	2.253
0.4043	0.9748	2.040	0.8388	0.9825	2.296
0.4295	0.9752	2.059	0.8926	0.9856	2.340
0.4497	0.9755	2.071	0.9503	0.9914	2.415
0.4793	0.9760	2.091	1.0000	1.0000	2.496
1-Butanol (1) + Cyclohexane (2)					
0.0000	0.0000	24.634	0.5014	0.0792	21.820
0.0276	0.0265	24.833	0.5493	0.0848	21.100
0.1002	0.0506	24.658	0.5497	0.0848	21.105
0.1495	0.0549	24.471	0.5991	0.0913	20.308
0.1988	0.0572	24.306	0.6002	0.0914	20.300
0.2495	0.0594	24.038	0.6484	0.0987	19.332
0.2990	0.0619	23.722	0.7006	0.1083	18.161
0.3491	0.0652	23.359	0.7504	0.1205	16.842
0.3988	0.0691	22.966	0.8001	0.1383	15.216
0.3995	0.0692	22.935	0.8513	0.1680	13.086
0.4487	0.0737	22.459	0.8972	0.2168	10.538
0.4496	0.0738	22.417	0.9533	0.3674	6.539
0.5001	0.0791	21.832	1.0000	1.0000	2.507
1-Butanol (1) + Methylbenzene (2)					
0.0000	0.0000	7.877	0.4991	0.2151	7.767
0.0491	0.0842	8.297	0.5487	0.2291	7.599
0.1441	0.1384	8.394	0.5494	0.2293	7.588
0.1804	0.1479	8.376	0.5987	0.2450	7.377
0.2213	0.1564	8.347	0.5993	0.2452	7.381
0.2591	0.1635	8.305	0.6497	0.2638	7.113
0.2984	0.1707	8.251	0.6983	0.2855	6.799
0.3479	0.1802	8.167	0.7493	0.3143	6.402
0.3987	0.1908	8.064	0.7988	0.3521	5.933
0.3987	0.1909	8.048	0.8435	0.4000	5.418
0.4488	0.2023	7.930	0.8935	0.4822	4.698
0.4490	0.2024	7.938	0.9491	0.6493	3.692
0.4986	0.2150	7.786	1.0000	1.0000	2.492

values of the different models which lead to the correlated results using Barker's method; the root-mean-square (rms) of the difference between the experimental and the calculated pressures (rms ΔP) and the maximum value of these pressure residuals (max ΔP) both are indicators of the quality of the agreement with data.

As an example, Figure 1 displays the plot of $(P_{\text{exp}} - P_{\text{calc}})$ against x_1 for the binary system (1-butanol (1) + *n*-heptane (2)), where the pressures were calculated using the Margules equation. We can see that, in the entire range of composition, there is a good agreement between experimental and calculated pressures. The same behavior has been observed for the other investigated binary mixtures.

■ DISCUSSION

The Margules equation leads to the best correlation results for the binary systems measured in this work: the rms deviation of the pressure is 0.060 kPa for (1-butanol + heptane), 0.053 kPa for (1-butanol + 2,2,4-trimethylpentane), 0.007 kPa for (1-butanol + dodecane), 0.078 kPa for (1-butanol + cyclohexane), and 0.014 kPa for (1-butanol + methylbenzene). The Margules equation gives also a maximum deviation varying between (0.013 and 0.176) kPa. The other models give higher values of the rms deviations for the majority of the systems. Our experimental vapor pressures for (1-butanol + methylbenzene) system at T = 313.15 K are in good agreement with values reported by Oracz et al.;[13] the average difference is 0.05 kPa. For the other binary systems, no experimental values were found for comparison.

All binary systems studied show an azeotrope except 1-butanol + dodecane as exposed in Figures 2 and 3. The azeotropic molar fractions of 1-butanol (x_1 azeotropic) and the azeotropic pressure (P azeotropic) at T = 313.15 K are reported in Table 3. The systems present a maximum pressure azeotrope, and the highest effect is obtained for the mixture containing cyclohexane.

Table 3. Parameters of the Models Used for the Binary Systems at T = 313.15 K together with the Root-Mean-Square Deviation of Pressure (rms ΔP), Maximum Value of Deviation (max ΔP), Azeotropic Mole Fractions of 1-Butanol ($x_{1,az}$), and Azeotropic Pressure (P_{az})

	Margules	Wilson	NRTL	UNIQUAC
1-Butanol (1) + Heptane (2)				
A_{12}	2.7580	0.0978	1.1059	1.3524
A_{21}	1.6782	0.4303	2.1528	0.3213
λ_{12}	2.3565			
λ_{21}	0.4973			
α_{12}			0.5139	
rms ΔP/kPa	0.060	0.060	0.065	0.245
max ΔP/kPa	0.130	0.130	0.138	0.414
$x_{1,az}$	0.0958	0.0958	0.0958	0.0958
P_{az}/kPa	13.125	13.106	13.112	12.833
1-Butanol (1) + 2,2,4-Trimethylpentane (2)				
A_{12}	2.6299	0.1217	1.1368	1.4120
A_{21}	1.7169	0.4280	2.0487	0.3105
λ_{12}	2.1417			
λ_{21}	0.6588			
α_{12}			0.5338	
rms ΔP/kPa	0.053	0.080	0.050	0.265
max ΔP/kPa	0.114	0.139	0.109	0.400
$x_{1,az}$	0.0981	0.0981	0.0981	0.0981
P_{az}/kPa	13.628	13.586	13.627	13.331
1-Butanol (1) + Dodecane (2)				
A_{12}	2.5537	0.1656	1.6199	1.4224
A_{21}	2.2263	0.2367	1.8469	0.3069
λ_{12}	1.8413			
λ_{21}	0.9643			
α_{12}			0.5037	
rms ΔP/kPa	0.007	0.016	0.008	0.065
max ΔP/kPa	0.013	0.041	0.019	0.186
1-Butanol (1) + Cyclohexane (2)				
A_{12}	2.5661	0.1054	0.8881	1.2943
A_{21}	1.3580	0.6190	2.1291	0.3767
λ_{12}	2.4163			
λ_{21}	0.4339			
α_{12}			0.5840	
rms ΔP/kPa	0.078	0.177	0.083	0.395
max ΔP/kPa	0.176	0.318	0.267	0.666
$x_{1,az}$	0.0276	0.0276	0.0276	0.0276
P_{az}/kPa	24.712	24.720	24.751	24.544
1-Butanol (1) + Methylbenzene (2)				
A_{12}	2.1336	0.1658	0.7290	1.2508
A_{21}	1.2603	0.6555	1.7005	0.4344
λ_{12}	1.5214			
λ_{21}	0.4156			
α_{12}			0.6107	
rms ΔP/kPa	0.014	0.028	0.013	0.066
max ΔP/kPa	0.037	0.052	0.032	0.117
$x_{1,az}$	0.1441	0.1441	0.1441	0.1441
P_{az}/kPa	8.381	8.357	8.381	8.301

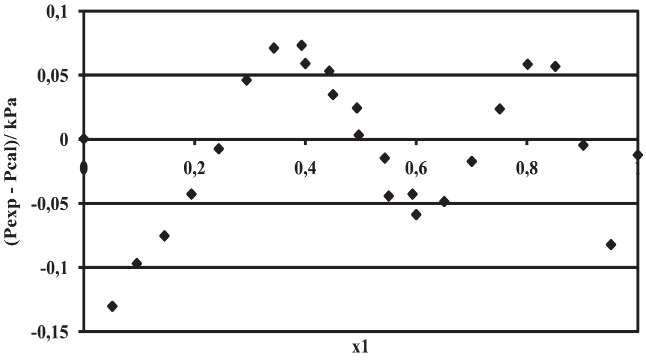

Figure 1. Pressure residuals ($P_{exp} - P_{calc}$) defined as differences between experimental and calculated pressures as a function of the liquid composition (x_1) of the system 1-butanol (1) + heptane (2).

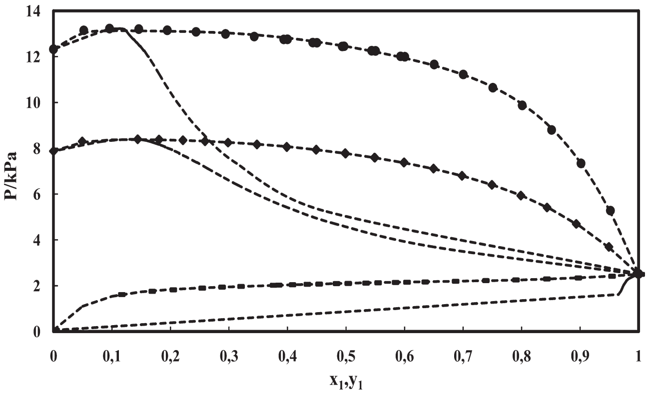

Figure 2. Total pressure at T = 313.15 K of the binary systems as a function of the liquid (x_1) and vapor composition (y_1): ●, 1-butanol (1) + heptane (2); —, 1-butanol (1) + dodecane (2); ◆, 1-butanol (1) + methylbenzene (2). Symbols represent the experimental points; lines are the calculations using the Margules equation.

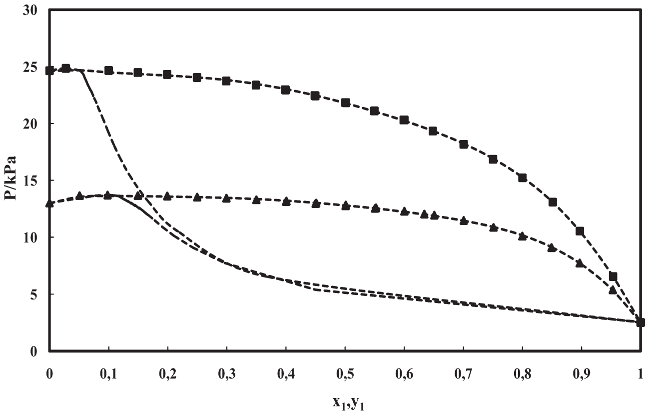

Figure 3. Total pressure at T = 313.15 K of the binary systems as a function of the liquid (x_1) and vapor composition (y_1): ▲, 1-butanol (1) + 2,2,4-trimethylpentane (2); ■, 1-butanol (1) + cyclohexane (2). Symbols represent the experimental points; lines are the calculations using the Margules equation.

Finally, we have calculated the excess molar Gibbs energy (G^E) for the binary systems. Figure 4 shows the values of G^E, calculated by the Margules equation, as a function of the liquid

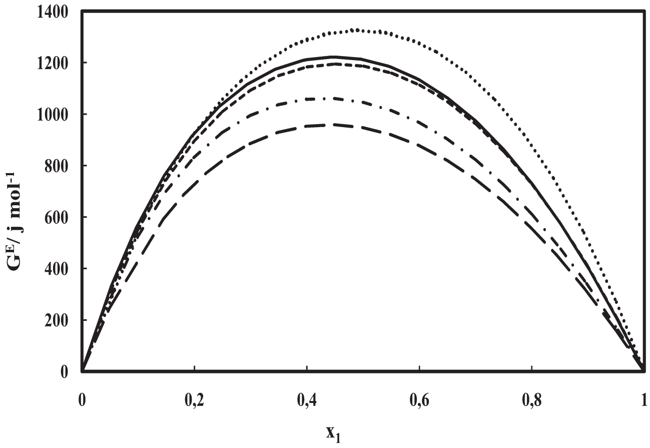

Figure 4. Excess molar Gibbs energy calculated using the Margules equation as a function of the molar fraction of the liquid phase (x_1) for binary systems: —, 1-butanol (1) + heptane (2); - - -, 1-butanol (1) + 2,2,4-trimethylpentane (2); ···, 1-butanol (1) + dodecane; - · -, 1-butanol (1) + cyclohexane; — —, 1-butanol (1) + methylbenzene (2).

mole fraction. The five binary systems measured exhibit a positive deviation from ideality. As expected, this effect is stronger for 1-butanol (1) + dodecane (2). Indeed, the G^E values for the composition of 1-butanol $x_1 \approx 0.50$ vary with the change of the chain of hydrocarbon: 1327 J·mol^{-1} (dodecane) > 1222 J·mol^{-1} (heptane) > 1195 J·mol^{-1} (2,2,4-trimethylpentane) > 1060 J·mol^{-1} cyclohexane) > 958 J·mol^{-1} (methylbenzene).

■ CONCLUSION

VLE data for the binary mixtures containing 1-butanol and hydrocarbons were measured at $T = 313.15$ K using a static device. The binary systems have been correlated by the Margules equation. Also the Wilson, NRTL, and UNIQUAC models were used for fitting the binary systems. Good results are obtained in the prediction of the total pressure for these systems.

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