Isobaric Vapor−Liquid Equilibria of Hexane + 1-Decene and Octane + 1-Decene Mixtures

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In this work the isobaric vapor−liquid equilibrium (VLE) data for hexane + 1-decene and octane + 1-decene systems have been measured at 101.33 kPa. The results were correlated using the Margules, Van Laar, Wilson, and UNIQUAC equations for the liquid phase activity coefficients. The average deviations fall within the same range for all models and agree well with experimental VLE data obtained in this work. Also, the experimental results were compared with the predictions of the modified UNIFAC group contribution method, which suggests that UNIFAC works well for unbranched alkane + 1-alkene systems.

Introduction

Reliable experimental vapor−liquid equilibrium (VLE) data are of both theoretical and practical significance in science and industry. From the scientific point of view, these data can be used to compare the strengths and weaknesses of various theoretical models that have been devised for this purpose, and on the other hand, in practice they are essential for the proper design of separation units for the purification of final products of an industrial chemical process.

Fischer−Tropsch synthesis is an important petrochemical process by means of which important categories of various chemicals ranging from low to high molecular weight saturated and unsaturated hydrocarbons may be produced.[1] There exists a large number of isothermal and isobaric experimental VLE data in the literature for *n*-alkane−1-alkene binary and ternary mixtures ranging from atmospheric to high-pressure conditions (see, for example, ref 2). However, in order to gain insight into the regular behavior of the phase equilibrium of these systems when one or both of the components change molecular weight (in a homologous series), it is necessary to have a more complete compilation of reliable experimental data.

The binary system hexane + 1-hexene has been widely studied[3–5] as well as hexane, 1-hexene, and octane binary and ternary systems[6] and the heptane + 1-hexene binary system.[7] Heptane + 1-heptene,[8–10] octane + 1-heptene,[9] and octane + 1-octene[11] are the other *n*-alkane + 1-alkene mixtures which have been studied to date. This work was undertaken as a part of an ongoing project dealing with the separation of mixtures formed by *n*-alkanes and 1-alkenes from the Fischer−Tropsch process. In this article, the vapor−liquid phase equilibria of the binary systems hexane + 1-decene and octane + 1-decene at a constant pressure of 101.33 kPa are studied by a modified Scott-ebulliometer, and the results are modeled by the correlative activity coefficient models, Margules, Van Laar, Wilson, and UNIQUAC, and also compared with the predictions of the UNIFAC method. The only data available in the literature for these systems are the infinite dilution activity coefficients, γ^{∞}, for hexane in 1-decene and octane in 1-decene at 298.15 K by Eckert, Carr, and co-workers.[12]

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

	ρ/kg·m^{-3} (293.15 K)		n_D (293.15 K)		T_b/K	
component	exptl	lit.	exptl	lit.	exptl	lit.
hexane	0.660	0.6606[15]	1.37486	1.3749[15]	341.80	341.85[14]
octane	0.708	0.7028[15]	1.39650	1.3974[15]	398.96	398.85[16]
1-decene	0.741	0.7408[15]	1.42140	1.4210[14]	443.80	443.65[14]

Experimental Section

Materials. 1-Decene and octane were supplied by Merck with nominal purities (~95 and >99) %, respectively, and hexane was supplied by BDH with nominal purity > 99 %. Octane and hexane were distilled and stored under nitrogen, and 1-decene was purified by the procedure described by Strukul & Michelin for the purification of 1-octene.[13] In this way, 1-decene was fractionally distilled under nitrogen atmosphere over sodium, which removed water and peroxides, and then stored under nitrogen. The purity of 1-decene was checked by gas chromatography to be 99.6 %. The measured refractive indices and boiling points of the chemicals used together with literature values[14–16] are listed in Table 1.

Apparatus and Procedure. In this work, isobaric data on vapor–liquid equilibria were obtained in a modified Scott-ebulliometer. Modification was made in such a way that dynamic recirculation of both liquid and vapor phases can be accomplished (Figure 1). The all-glass recirculation still consists of a 175 mL reflux and mixing chamber equipped with a Cottrell tube, an equilibrium chamber, a liquid sampling valve, and a vapor-sampling valve. The apparatus is capable of handling pressures from (1.00 to 101.33) kPa and temperatures up to 473.15 K. The Cottrell pump ensures that both liquid and vapor phases are in intimate contact during boiling and also in contact with the temperature sensing element. The mixture of components is heated and mixed in the reflux chamber by means of a heater-stirrer and a magnetic bar inside the reflux chamber. The reflux chamber, Cottrell pump, and equilibrium chamber are enclosed by a sealed evacuated glass shield, which in turn is

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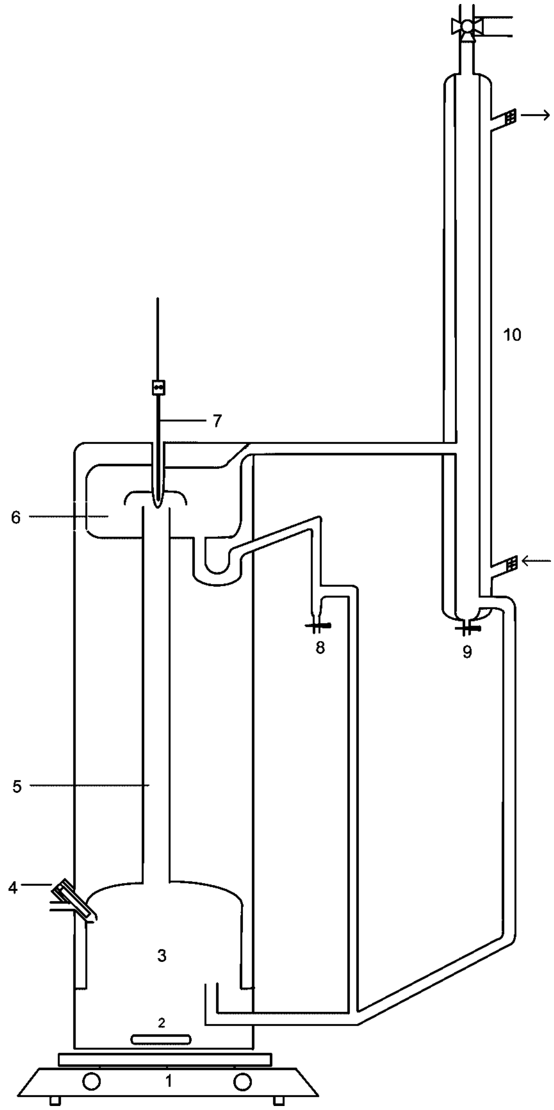

Figure 1. Modified Scott-ebulliometer: 1, heater-stirrer; 2, magnetic bar; 3, reflux and mixing still; 4, sample inlet valve; 5, Cottrell pump; 6, equilibrium tank; 7, Pt-100 sensor; 8, liquid sampling valve; 9, vapor sampling valve; 10, condenser.

wrapped by a thermal insulating jacket (not shown in Figure 1) to prevent loss of heat from the system to the surroundings.

The equilibrium temperature was measured with a Lutron model TM-917 digital thermometer with a 0.01 K resolution using a Pt-100 sensor positioned at the Cottrell pump shown in Figure 1. The pressure of the system was measured and controlled by the setup shown in Figure 2. In each experiment, the pressure was fixed to 101.33 kPa and was read to ± 0.06 kPa. For this purpose, at first the apparatus was evacuated with a vacuum pump, and then about 175 mL of the liquid mixture, which was initially placed under nitrogen atmosphere in a sealed bottle, was transferred to the still via a sample inlet valve. The liquid was heated to boiling, and the pressure of the system was adjusted to 101.33 kPa by introducing nitrogen gas through the drying vessel (5) in Figure 2. The still was operated under constant pressure until equilibrium was reached normally after about 30 min. Heating of the mixture was regulated to achieve a mean speed of 25 to 30 drops per min. Equilibrium conditions were assumed when constant temperature and pressure were obtained for 30 min. Samples of 0.5 mL were taken simultaneously from the liquid and condensed vapor, and their compositions were analyzed with a calibrated Bellingham & Stanly RFM91 refractometer at 10 °C (to prevent loss of the more volatile component) to within 0.00001. The uncertainty of the measured mole fraction was ± 0.0001.

Results and Discussion

The accuracy of the instrument employed was checked by measuring the isobaric VLE of the binary mixture of the ethanol + 1-octanol system at 101.33 kPa and compared with the existing reliable data in the literature.[17] The *Txy* phase diagram of this system presented in Figure 3 shows good agreement between the data measured in this work and the experimental values of Arce et al.[17]

Experimental data for the vapor−liquid equilibrium of the hexane + 1-decene and octane + 1-decene systems at 101.33 kPa together with the liquid phase activity coefficients of both components are presented in Tables 2 and 3, respectively. The activity coefficients fluctuate smoothly around 1, showing, on average, a slightly positive deviation from ideal behavior except for the activity coefficient of 1-decene in the octane + 1-decene mixture, which shows a slightly negative deviation from ideal behavior. The activity coefficients have been calculated according to the thermodynamics equilibrium criteria. The Poynting factor was assumed equal to unity since the operating pressure was atmospheric (the values of the Poynting factor vary smoothly from 0.955 to 1.007 and from 0.989 to 1.005 for hexane + 1-decene and octane + 1-decene mixtures, respectively, using the Spencer and Danner modification[18] of the Rackett equation for estimating the saturated volumes of pure components). The virial equation used was truncated after the first two terms. The applied equations are as follows

$$y_iP = x_i\gamma_iP_i^{\mathrm{s}}\exp\left[-\frac{B_{ii}(P-P_i^{\mathrm{s}})+(1-y_i)^2P\delta_{ij}}{RT}\right] \qquad (1)$$

$$\delta_{ij} = 2B_{ij} - B_{ii} - B_{jj}$$

This comes from

$$y_i\Phi_iP = x_i\gamma_iP_i^{\mathrm{s}} \qquad (2)$$

where

$$\Phi_i = \exp\left[\frac{B_{ii}(P-P_i^{\mathrm{s}})+(1-y_i)^2P\delta_{ij}}{RT}\right] \qquad (3)$$

where γ_i is the activity coefficient of component i; x_i and y_i are the equilibrium mole fractions of component i in the liquid and vapor phases; P is the total pressure; P_i^{s} is the saturated vapor pressure of component i; R is the gas constant; T is the absolute temperature; and B_{ii} and B_{ij} are the pure-component and cross second virial coefficients, respectively, which have been calculated using the Pitzer and Curl equations with the corrections proposed by Tsonopoulos[19] and mixing rules of Prausnitz.[20] The binary interaction parameter, k_{12}, was estimated by the relationship proposed by Chueh and Prausnitz[21] for nonpolar pairs being 0.00884 for hexane + 1-decene and 0.00140 for octane + 1-decene systems, respectively.

The Antoine equation was used to calculate the vapor pressures of the pure compounds, the parameters of which were obtained by fitting the Antoine equation to experimental vapor pressure data[14] and are summarized in Table 4.

After obtaining the activity coefficients from the experimental VLE data (eqs 1 and 2), several models were used for correlation. Equations based on classic models (Margules[22] and Van Laar[23]) and two models based on the local composition concept (Wilson[24] and UNIQUAC[25]) were applied. To obtain

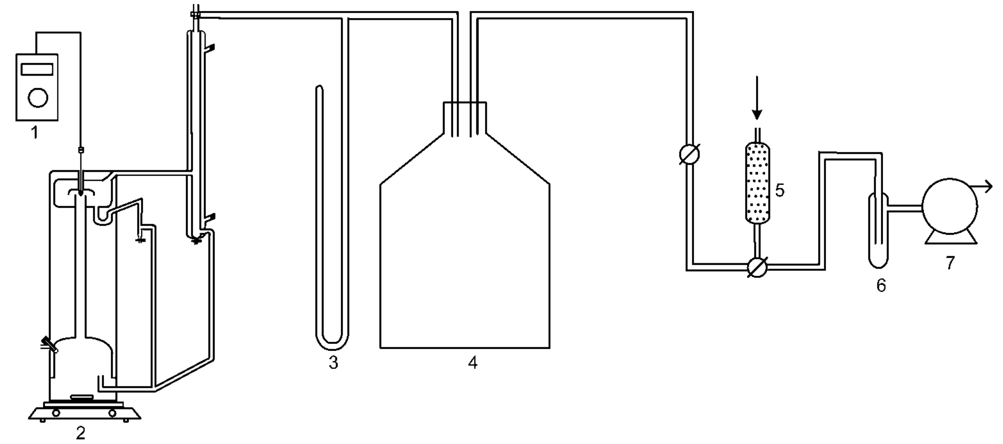

Figure 2. System for the determination of vapor–liquid equilibria: 1, temperature read-out device; 2, modified Scott-ebulliometer; 3, mercury manometer; 4, 5 L ballast bulb for pressure control; 5, drying vessel ($CaCl_2$); 6, trap; 7, vacuum pump.

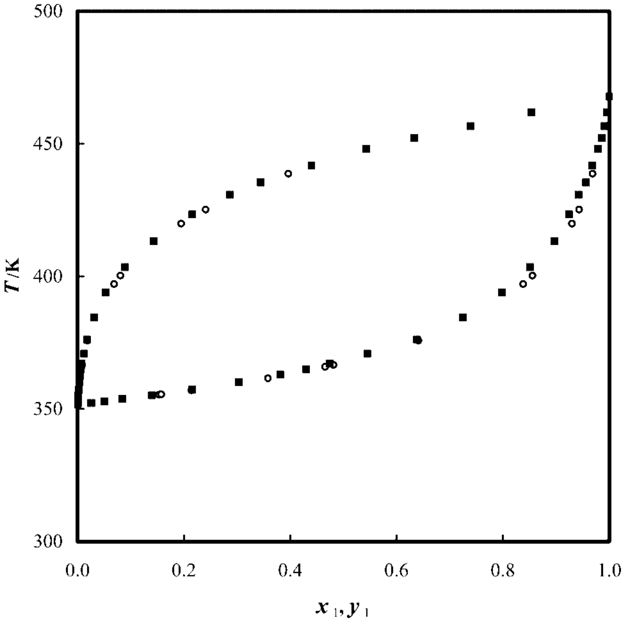

Figure 3. VLE (*Txy*) of the system 1-octanol (1) + ethanol (2) at 101.33 kPa pressure: ○, this work; and ■, from ref 17.

the binary interaction parameters characteristic of the different equations, the following objective function (OF) was minimized

$$\mathrm{OF} = \sum (\gamma_{1,\mathrm{exptl}} - \gamma_{1,\mathrm{calcd}})^2 + \sum (\gamma_{2,\mathrm{exptl}} - \gamma_{2,\mathrm{calcd}})^2 \quad (4)$$

where $\gamma_{i,\mathrm{exptl}}$ and $\gamma_{i,\mathrm{calcd}}$, are experimental and calculated activity coefficient of component *i* in the binary mixture, respectively. This multiparametric regression was carried out by means of the SOLVER macro function of the Excel Microsoft Office 1997 data calculation sheet. The values of the adjusted parameters, A_{12} and A_{21}, together with the average deviations obtained by comparing the calculated values from different correlations to the experimental data are shown in Table 5. It can be seen that the average deviations fall within the same range for all models studied in this work. From the data in Table 5, it can be concluded that all of the theoretical models agree well with VLE data obtained in this work. This behavior may be ascribed to the nonpolar nature of the components of the mixtures studied in this work in which their behavior can be predicted equally well by the models considered here. For example, Figures 4 and 5 illustrate the good agreement between the experimental and calculated data using the UNIQUAC equation. Also Table 5 shows that uncertainties in the data are greater for hexane + 1-decene than for octane + 1-decene. This is because the difference between the boiling points of components is higher for hexane + 1-decene than for octane + 1-decene, which gives rise to higher uncertainty in the measurement of temperature of the mixture.

Table 2. Boiling Temperatures *T*, Liquid and Vapor Phase Mole Fractions x_1, y_1, and Activity Coefficients for Hexane (1) + 1-Decene (2) at 101.33 kPa

x_1	y_1	T/K	γ_1	γ_2
		Hexane (1) + 1-Decene (2)		
0.0000	0.0000	443.80		1.0000
0.0152	0.1317	438.51	1.0628	1.0037
0.0417	0.3346	429.68	1.1304	0.9929
0.0644	0.4603	423.04	1.1243	0.9876
0.0830	0.5402	418.14	1.1149	0.9849
0.1091	0.6278	411.95	1.1016	0.9828
0.1476	0.7203	404.04	1.0849	0.9815
0.1795	0.7751	398.37	1.0737	0.9813
0.2173	0.8230	392.50	1.0626	0.9813
0.2676	0.8682	385.81	1.0502	0.9814
0.3046	0.8922	381.58	1.0422	0.9815
0.3610	0.9192	375.97	1.0307	0.9820
0.4133	0.9372	371.53	1.0204	0.9833
0.4553	0.9482	368.39	1.0121	0.9855
0.5047	0.9584	365.10	1.0023	0.9903
0.5356	0.9637	363.22	0.9960	0.9949
0.5919	0.9715	359.91	0.9902	1.0172
0.6700	0.9796	355.62	0.9889	1.0771
0.7628	0.9864	351.39	0.9826	1.1939
0.8250	0.9899	348.52	0.9885	1.3695
1.0000	1.0000	341.80	1.0000	

The thermodynamic consistency of the experimental data was checked by means of a modified Dechema test,[26] where the fugacity coefficients are calculated by the method of Hayden and O'Connell[27] and activity coefficients are calculated by using the four-suffix Margules equation

$$g^{\mathrm{E}}/RT = x_1x_2(Ax_2 + Bx_1 - Dx_1x_2) \quad (5)$$

with the corresponding activity coefficients

$$\ln \gamma_1 = x_2^2[A + 2(B - A - D)x_1 + 3Dx_1^2] \quad (6)$$

$$\ln \gamma_2 = x_1^2[B + 2(A - B - D)x_2 + 3Dx_2^2] \quad (7)$$

Parameters *A*, *B*, and *D* were estimated by the method described previously for the theoretical models. The values of the parameters *A*, *B*, and *D* together with the average absolute deviation, Δy_1, for the two systems investigated in this work are presented in Table 6. To pass the consistency test, a system

Table 3. Boiling Temperatures T, Liquid and Vapor Phase Mole Fractions x_1, y_1, and Activity Coefficients for Octane (1) + 1-Decene (2) at 101.33 kPa

x_1	y_1	T/K	γ_1	γ_2
Octane (1) + 1-Decene (2)				
0.0000	0.0000	443.80		1.0000
0.0182	0.0471	442.37	0.9557	1.0040
0.0402	0.1075	440.69	1.0203	1.0026
0.0547	0.1446	439.61	1.0308	1.0020
0.0614	0.1601	439.12	1.0271	1.0030
0.0899	0.2272	437.09	1.0373	1.0016
0.1157	0.2833	435.32	1.0422	0.9999
0.1568	0.3645	432.64	1.0464	0.9963
0.1720	0.3922	431.69	1.0473	0.9948
0.2129	0.4610	429.22	1.0486	0.9902
0.2607	0.5321	426.51	1.0485	0.9841
0.2902	0.5714	424.92	1.0479	0.9801
0.3288	0.6184	422.92	1.0466	0.9746
0.3323	0.6225	422.74	1.0464	0.9741
0.3715	0.6650	420.82	1.0445	0.9685
0.3997	0.6930	419.50	1.0428	0.9645
0.4325	0.7231	418.00	1.0406	0.9600
0.4517	0.7395	417.16	1.0392	0.9574
0.4862	0.7669	415.69	1.0366	0.9532
0.4975	0.7754	415.22	1.0357	0.9519
0.5013	0.7782	415.06	1.0353	0.9515
0.5307	0.7990	413.88	1.0328	0.9485
0.5657	0.8218	412.51	1.0297	0.9458
0.6030	0.8439	411.11	1.0262	0.9440
0.6210	0.8539	410.45	1.0244	0.9438
0.6691	0.8785	408.76	1.0195	0.9455
0.6935	0.8900	407.93	1.0170	0.9482
0.7201	0.9016	407.06	1.0142	0.9530
0.7555	0.9161	405.92	1.0104	0.9634
0.7810	0.9257	405.13	1.0076	0.9750
0.7817	0.9260	405.11	1.0075	0.9754
0.7889	0.9286	404.89	1.0067	0.9795
0.8398	0.9459	403.38	1.0010	1.0243
0.8978	0.9665	401.74	0.9977	1.0471
0.9618	0.9891	400.03	0.9962	0.9627
1.0000	1.0000	398.96	1.0000	

Table 4. Antoine Equation Parameters for the Pure Components[a]

component	A	B/K	C/K
hexane	6.4723	1208.11	201.83
octane	5.5253	1145.87	199.76
1-decene	6.2947	1642.34	212.28

[a] $\log_{10} P_{\mathrm{vp}}/\mathrm{kPa} = A - B/((T/\mathrm{K}) + C - 273.15)$.

Table 5. Values of the Parameters Used for Each Model and Average of Absolute Deviations (Δy, ΔT) between Calculated and Experimental Data for the Vapor Phase Mole Fraction and Temperature

model	A_{12}	A_{21}	Δy_1[a]	ΔT/K[a]
Hexane (1) + 1-Decene (2)				
Margules	−0.06062	−0.05567	0.00342	0.535
Van Laar	−0.06073	−0.05571	0.00342	0.535
Wilson	0.55690	1.64556	0.00340	0.540
UNIQUAC	0.77159	1.26751	0.00339	0.538
Octane (1) + 1-Decene (2)				
Margules	0.01893	0.01203	0.00318	0.046
Van Laar	0.01945	0.01254	0.00319	0.048
Wilson	0.75194	1.26577	0.00318	0.048
UNIQUAC	0.90261	1.09682	0.00318	0.047

[a] $\Delta X = \sum_{i=1}^{N} |X_{i,\mathrm{exptl}} - X_{i,\mathrm{calcd}}| / N$.

must have an average deviation less than 0.01. The two systems included in this work have passed this consistency test.

Finally, the activity coefficients for two systems investigated in this work have been calculated by the UNIFAC group contribution method.[28,29] The average absolute deviations between experimental data and calculated values by the UNIFAC method are summarized in Table 7.

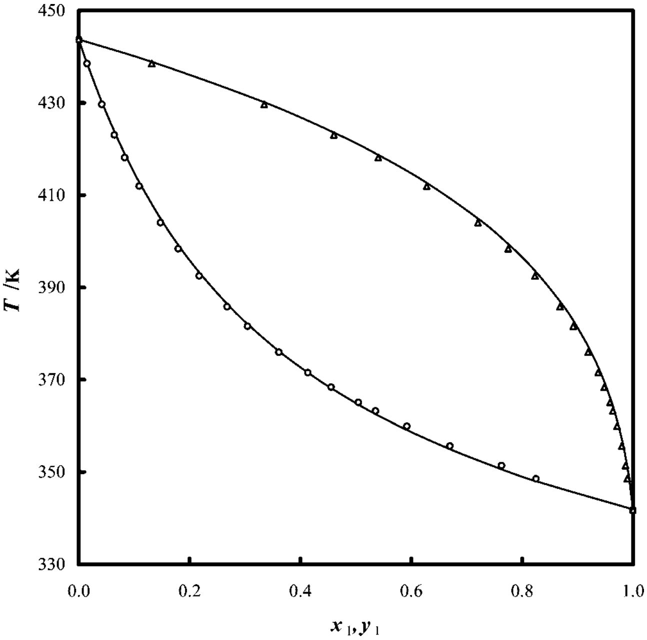

Figure 4. VLE (Txy) of the system hexane (1) + 1-decene (2) at 101.33 kPa pressure: ○, Δ, experimental values; −, the UNIQUAC model.

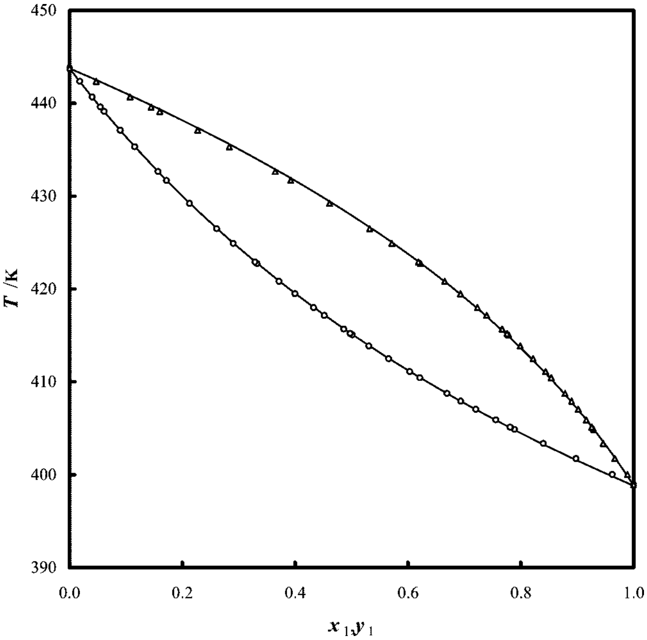

Figure 5. VLE (Txy) of the system octane (1) + 1-decene (2) at 101.33 kPa pressure: ○, Δ, experimental values; −, the UNIQUAC model.

Table 6. Results of the Thermodynamic Consistency Test

system	A	B	D	Δy_1
hexane (1) + 1-decene (2)	−0.06114	−0.05613	−0.00266	0.00343
octane (1) + 1-decene (2)	0.01896	0.01205	0.00014	0.00318

Conclusions

New isobaric vapor–liquid equilibrium data not previously reported in the literature have been measured for the systems hexane + 1-decene and octane + 1-decene. Both systems show a near ideal behavior. The binary parameters of four activity coefficient models, namely, Margules, Van Laar, Wilson, and UNIQUAC, were evaluated for the binary systems studied in this work. Results show experimental VLE data are thermodynamically consistent, and there is excellent agreement between models and experimental data. Also the experimental results

Table 7. Average Absolute Deviations between Experimental Equilibrium *y* and *T* Data and Data Obtained by Using the UNIFAC Method

mixture	Δy_1	ΔT/K
hexane (1) + 1-decene (2)	0.0034	0.525
octane (1) + 1-decene (2)	0.0032	0.049

confirm the VLE data values generated by the predictive UNIFAC group contribution method. This suggests that for engineering purposes the UNIFAC method may be used for any other unbranched alkane + alkene system in the C5 to C11 range when experimental data do not exist.

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