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Isobaric Vapor–Liquid Equilibria at 97.3 kPa and Excess Properties at (303.15, 308.15, and 313.15) K of Binary Mixture of *p*-Xylene + Decane

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Isobaric vapor–liquid equilibrium data were obtained for the system *p*-xylene + decane at 97.3 kPa using a vapor recirculating type (modified Othmer's) equilibrium still. The activity coefficients were obtained by taking into consideration the vapor-phase nonideality. A comparison of values of activity coefficients obtained by experimental data was made with UNIFAC. The experimental data for the system were tested for thermodynamic consistency and correlated by various equations. Also the excess molar volumes, viscosities, and speeds of sound for the binary mixture at $T = (303.15, 308.15, \text{ and } 313.15) \text{ K}$ had been measured over the whole compositional range. The excess thermodynamic properties such as deviations in viscosity ($\Delta\eta$) and excess isentropic compressibility (K_S^E) were calculated.

Introduction

In recent years, there has been considerable upsurge in the theoretical and experimental investigations of the vapor–liquid equilibria and excess thermodynamic properties of binary and multicomponent mixtures. The physical, chemical, and thermodynamic properties associated with the liquids and liquid mixtures such as excess molar volume, viscosity, compressibility, vapor–liquid equilibria, and liquid–liquid equilibria directly depend on the nature of the molecules that constitute it, so a thorough knowledge of molecular behavior is therefore essential to understand completely the physical or chemical behavior of a substance. In principle, the interaction between the molecules can be established from a study of the characteristic abrupt departure from ideal behavior of some physical properties such as volume, compressibility, and viscosity. The study of these properties is used as a tool for investigating the properties of the mixture and the nature of intermolecular interactions between the components constituting the mixture.

We have recently reported studies on 2-propanol and 3-pentanone with *p*-xylene.¹ In our earlier studies, we have reported the thermodynamic behavior of several binary systems.^{2,3}

In the present work, isobaric vapor–liquid equilibrium data were obtained for *p*-xylene + decane system at 97.3 kPa using a modified version of the recirculating type equilibrium still described earlier.^{4,5} The system has a boiling range of 35.77 K and does not form an azeotrope.

Also in the present study, the experimental values of the excess molar volume, viscosity, and speed of sound at three temperatures, $T = (303.15, 308.15, \text{ and } 313.15) \text{ K}$, for the system *p*-xylene + decane have been determined over the entire compositional range. From the experimental data, densities, deviations in viscosity, and excess isentropic compressibility have been calculated. The densities and viscosities for this system at 298.15 K have already been reported earlier in the literature.⁶

Experimental Section

p-Xylene (Riedel, Germany) and decane (Merck, Schuchardt, Germany), AR grade, were purified using standard procedures⁷ and stored over molecular sieves. The purity of the chemicals was checked by comparing the measured densities and viscosities with those reported in the literature as shown in Table 1.

A modified version of the equilibrium still⁵ was used for obtaining the vapor–liquid equilibrium data. The equilibrated mixtures were analyzed by the refractive index method using a Bausch and Lomb Abbe-3L refractometer. The apparatus, modifications, and analytical techniques have already been described earlier.⁸ The estimated uncertainties in the measurements of mole fraction were ± 0.0002 , in temperature were $\pm 0.1 \text{ K}$, and in pressure were $\pm 0.27 \text{ kPa}$.

Excess molar volumes, which are reproducible to $\pm 0.003 \text{ cm}^3\cdot\text{mol}^{-1}$, were measured with a continuous dilution dilatometer.⁹ The density values have been estimated from the excess molar volume data with reproducibility better than $\pm 1 \times 10^{-4}$. Viscosities were measured with the help of modified Ubbelohde viscometer as described earlier.³ At each temperature, the viscometer was calibrated so as to determine the two constants A and B in the equation:

$$\eta/\rho = At + B/t \quad (1)$$

The values of constants were obtained by measuring the flow time (t) with triple-distilled water and double-distilled benzene. The flow measurements were made with an electronic stop watch with precision of $\pm 0.01 \text{ s}$. The reproducibility of viscosity results was found to be within $\pm 0.003 \text{ mPa}\cdot\text{s}$. Speeds of sound were measured with an interferometer (UTI-101) with an uncertainty of $\pm 0.1 \text{ m}\cdot\text{s}^{-1}$. The isentropic compressibility values estimated from the speed of sound data were within uncertainty of $\pm 0.2 \text{ T}\cdot\text{Pa}^{-1}$. All the measurements were made at a constant temperature with the help of a circulating type cryostat (Type MK70, MLW, Germany) maintained at a temperature within $\pm 0.02 \text{ K}$.

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Table 1. Physical Properties of the Pure Components at Different Temperatures

component	<i>T</i> /K	$\rho \times 10^{-3}/\text{kg}\cdot\text{m}^{-3}$		$\eta/\text{mPa}\cdot\text{s}$	
		expt	lit	expt	lit
<i>p</i> -xylene	303.15	0.8523	0.85230 ¹⁷	0.566	0.566 ⁷
	308.15	0.8479	0.84790 ¹⁸	0.537	0.539 ¹⁸
	313.15	0.8436	0.84364 ¹⁹	0.513	0.513 ²⁰
decane	303.15	0.7223	0.72238 ¹⁷	0.786	0.786 ²¹
	308.15	0.7182	0.71860 ²¹	0.717	0.717 ²²
	313.15	0.7147	0.71476 ²³	0.694	0.694 ²⁴

Table 2. Physical Constants of the Pure Components

constant	<i>p</i> -xylene	decane
molecular weight	106.169 ²⁵	142.287 ²⁵
boiling point/K (at 101.3 kPa)	411.501 ²⁵	447.273 ²⁵
refractive index (at 298.15 K)	1.49325 ²⁵	1.40967 ²⁵
<i>T</i> _g /K	616.2 ²⁶	617.7 ²⁶
<i>P</i> _c /kPa	3509.1 ²⁶	2119.5 ²⁶
<i>V</i> _c × 10 ⁶ /m ³ ·mol ^{−1}	379.00 ²⁶	603.00 ²⁶
acentric factor, ω	0.32 ²⁶	0.489 ²⁶
dipole moment, μ /debye	0 ²⁶	0 ²⁶
Antoine constants, eq 4		
<i>A</i>	6.98820 ²⁷	6.95707 ²⁷
<i>B</i>	1451.792 ²⁷	1503.568 ²⁷
<i>C</i>	215.111 ²⁷	194.738 ²⁷

Table 3. Vapor–Liquid Equilibrium Data of *p*-Xylene(1) + Decane(2) at 97.3 kPa

<i>T</i> /K	<i>x</i> ₁	<i>y</i> ₁	ln γ ₁	ln γ ₂
410.0	1.0000	1.0000	0.0000	0.2620
410.1	0.9900	0.9910	0.0001	0.2520
410.2	0.9740	0.9874	0.0020	0.2429
410.9	0.9531	0.9771	0.0030	0.2257
411.6	0.9230	0.9616	0.0030	0.2050
414.9	0.8221	0.8986	0.0040	0.1340
416.6	0.7579	0.8626	0.0041	0.1100
420.0	0.6390	0.7804	0.0139	0.0769
421.2	0.5866	0.7474	0.0296	0.0640
423.2	0.4945	0.6841	0.0594	0.0601
425.9	0.4031	0.6097	0.0800	0.0505
427.8	0.3500	0.5592	0.0987	0.0379
430.6	0.2500	0.4706	0.1429	0.0300
431.8	0.2000	0.4281	0.1830	0.0283
433.7	0.1500	0.3738	0.2278	0.0190
434.2	0.1363	0.3560	0.2454	0.0190
435.8	0.0850	0.2972	0.3127	0.0200
438.4	0.0450	0.2180	0.3787	0.0150
439.6	0.0338	0.1812	0.3950	0.0140
445.7	0.0000	0.0000	0.4560	0.0000

Table 4. Correlation Parameters for Activity Coefficient and Deviation in Vapor Phase Composition

correlations	<i>A</i> ₁	<i>A</i> ₂	<i>A</i> ₃	deviation ($\Delta\gamma$)
Margules	0.43559	0.27054	0.35417	0.0114
Redlich–Kister	0.26453	−0.08253	0.08854	0.0096
Black	−0.00163	0.00954	0.34113	0.1891

Results and Correlations

The liquid-phase activity coefficients (γ) were calculated from the experimental data using the equations below, which take into account the vapor-phase nonideality:

$$\gamma_1 = (Py_1/P_1^0 x_1) \exp[(B_{11} - V_1)(P - P_1^0)/RT + P\delta_{12}y_2^2/RT] \quad (2)$$

$$\gamma_2 = (Py_2/P_2^0 x_2) \exp[(B_{22} - V_2)(P - P_2^0)/RT + P\delta_{12}y_1^2/RT] \quad (3)$$

where *x* and *y* are the equilibrium mole fraction in the liquid and vapor phases, *T* and *P* are the boiling point and

Table 5. Excess Molar Volume (*V*^E), Viscosity (η), Speed of Sound (*u*), Density (ρ), and Compressibility (*K*_S) for *p*-Xylene (1) + Decane (2) at Different Temperatures

<i>x</i> ₁	<i>V</i> ^E × 10 ⁶ (m ³ ·mol ^{−1})	η (mPa·s)	<i>u</i> (m·s ^{−1})	$\rho \times 10^{-3}$ (kg·m ^{−3})	<i>K</i> _S (T·Pa ^{−1})
<i>T</i> = 303.15 K					
1.0000	0.000	0.566	1283.7	0.8523	712.0
0.9332	0.172	0.566	1274.4	0.8380	734.8
0.8604	0.273	0.568	1265.3	0.8241	758.0
0.7777	0.354	0.576	1255.4	0.8098	783.5
0.6938	0.426	0.588	1246.6	0.7966	807.9
0.6001	0.490	0.605	1237.9	0.7831	833.3
0.4949	0.529	0.628	1230.0	0.7695	859.0
0.4189	0.514	0.646	1225.5	0.7607	875.3
0.2952	0.435	0.680	1220.0	0.7477	898.6
0.1533	0.283	0.729	1215.1	0.7346	922.0
0.0000	0.000	0.786	1210.0	0.7223	945.6
<i>T</i> = 308.15 K					
1.0000	0.000	0.537	1271.0	0.8479	730.1
0.9332	0.303	0.537	1251.0	0.8328	767.3
0.8604	0.455	0.539	1238.4	0.8187	796.4
0.7777	0.556	0.546	1229.0	0.8043	823.1
0.6938	0.632	0.557	1221.6	0.7912	847.0
0.6001	0.703	0.572	1214.1	0.7778	872.3
0.4949	0.759	0.592	1207.3	0.7642	897.8
0.4189	0.760	0.609	1203.1	0.7554	914.6
0.2952	0.680	0.639	1197.6	0.7425	939.1
0.1533	0.489	0.678	1192.3	0.7296	964.1
0.0000	0.000	0.717	1189.0	0.7182	984.9
<i>T</i> = 313.15 K					
1.0000	0.000	0.513	1260.0	0.8436	746.6
0.9332	0.440	0.515	1232.8	0.8278	794.9
0.8604	0.647	0.519	1221.3	0.8134	824.2
0.7777	0.771	0.527	1212.5	0.7992	851.2
0.6938	0.876	0.538	1205.6	0.7860	875.3
0.6001	0.941	0.552	1198.8	0.7728	900.4
0.4949	0.981	0.573	1192.0	0.7595	926.7
0.4189	0.980	0.590	1187.7	0.7507	944.2
0.2952	0.911	0.623	1182.0	0.7379	969.9
0.1533	0.699	0.660	1176.9	0.7253	995.5
0.0000	0.000	0.694	1175.3	0.7147	1012.8

Table 6. Values of Coefficients of Redlich–Kister (Equation 10) and Standard Deviations (Equation 11)

<i>T</i> /K	<i>A</i> ₀	<i>A</i> ₁	<i>A</i> ₂	<i>A</i> ₃	σ
<i>V</i> ^E × 10 ⁶ /m ³ ·mol ^{−1}					
303.15	2.0626	−0.2977	0.2313	0.8283	0.011
308.15	2.9641	−0.6934	1.4898	1.5322	0.017
313.15	3.8535	−0.6474	2.9363	1.4138	0.025
$\Delta\eta/\text{mPa}\cdot\text{s}$					
303.15	−0.1992	−0.0169	−0.0154	−0.0394	0.0007
308.15	−0.1448	−0.0559	0.0184	−0.0322	0.0004
313.15	−0.1262	−0.0639	0.0485	−0.0310	0.0004
<i>K</i> _S ^E /T·Pa ^{−1}					
303.15	10.1573	−15.6679	−31.6172	10.7367	0.19
308.15	39.0000	10.0302	74.5714	93.1823	0.77
313.15	58.6326	0.6996	146.4497	182.1912	0.18

the total pressure, *V* is the molar liquid volume, *B*₁₁ and *B*₂₂ are the second virial coefficients of the pure components, *B*₁₂ is the cross second virial coefficient, and

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

The pure component vapor pressures (*P*⁰) were calculated according to the Antoine equation:

$$\log(0.133 P^0/\text{kPa}) = A - [B/((C - 273.15) + (T/K))] \quad (4)$$

where the constants *A*, *B*, and *C* are reported in Table 2. The Lyckman et al.¹⁰ correlation was used for the estimation of liquid molar volumes. The Pitzer and Curl

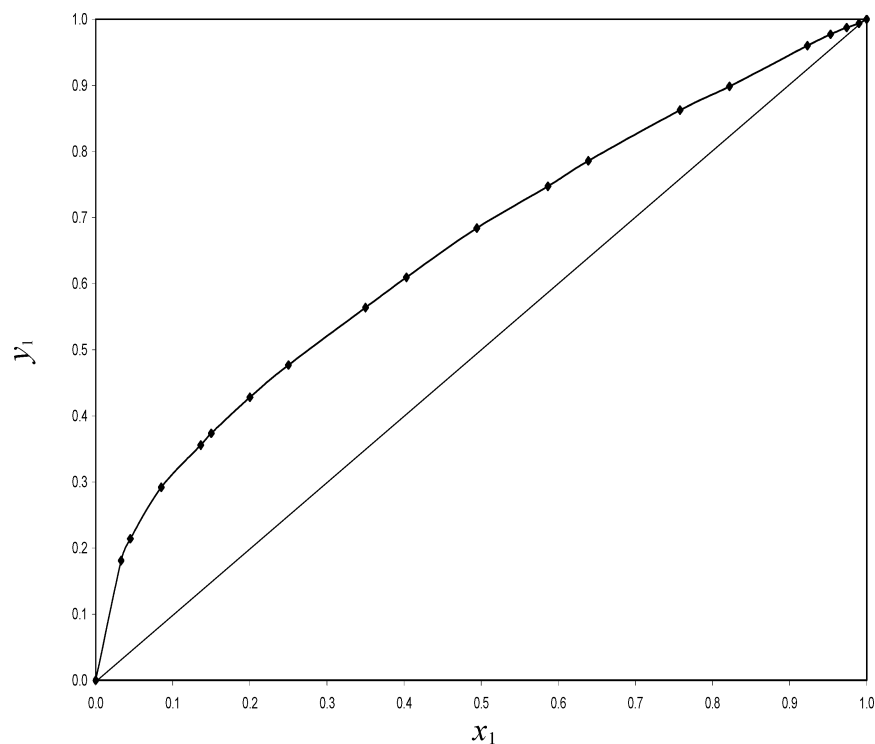


Figure 1. Plot of vapor–liquid equilibrium data for the system *p*-xylene (1) + decane (2) at 97.3 kPa.

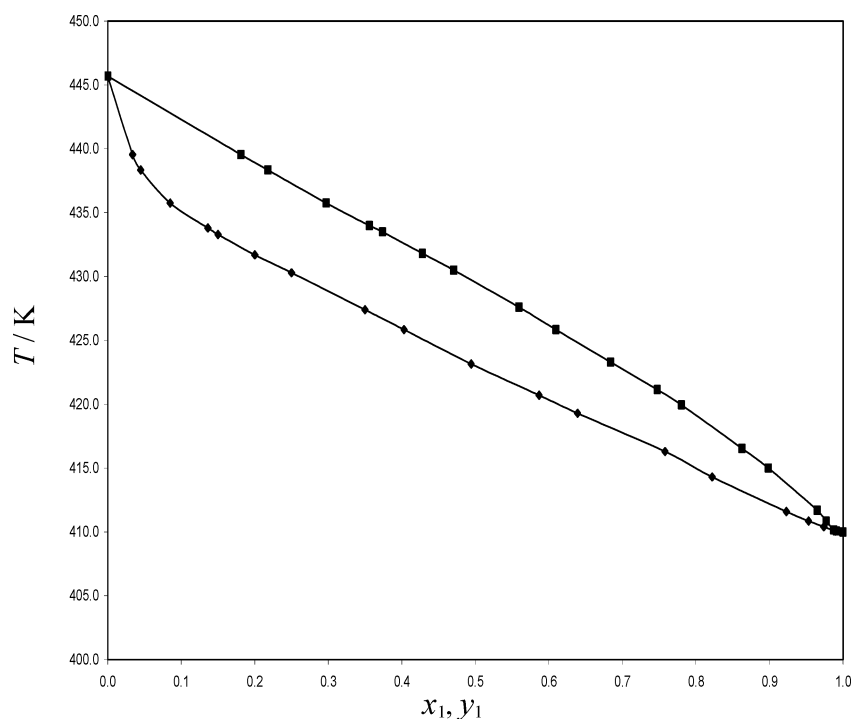


Figure 2. Plot of T vs x_1, y_1 data for the system *p*-xylene (1) + decane (2).

equation modified by Tsonopoulos¹¹ was used in the evaluation of cross virial coefficients in this work.

The experimental values of excess volume (V^E), viscosity (η), and speed of sound (u) measured at different temperatures for the system are listed in Table 5. The density of the binary mixture was calculated from the excess molar volume data by the following relation:

$$\rho_m = (M_1x_1 + M_2x_2)/(V^E + x_1V_1^0 + x_2V_2^0) \quad (5)$$

where x_1 and x_2 are mole fractions, M_1 and M_2 are molecular masses, and V_1^0 and V_2^0 are molar volumes of pure components 1 and 2, respectively.

The deviations in viscosity ($\Delta\eta$) were calculated by using

$$\Delta\eta = \eta_m - (x_1\eta_1 + x_2\eta_2) \quad (6)$$

where η_1 and η_2 are the viscosities of pure components 1 and 2, respectively, and η_m is the mixture viscosity.

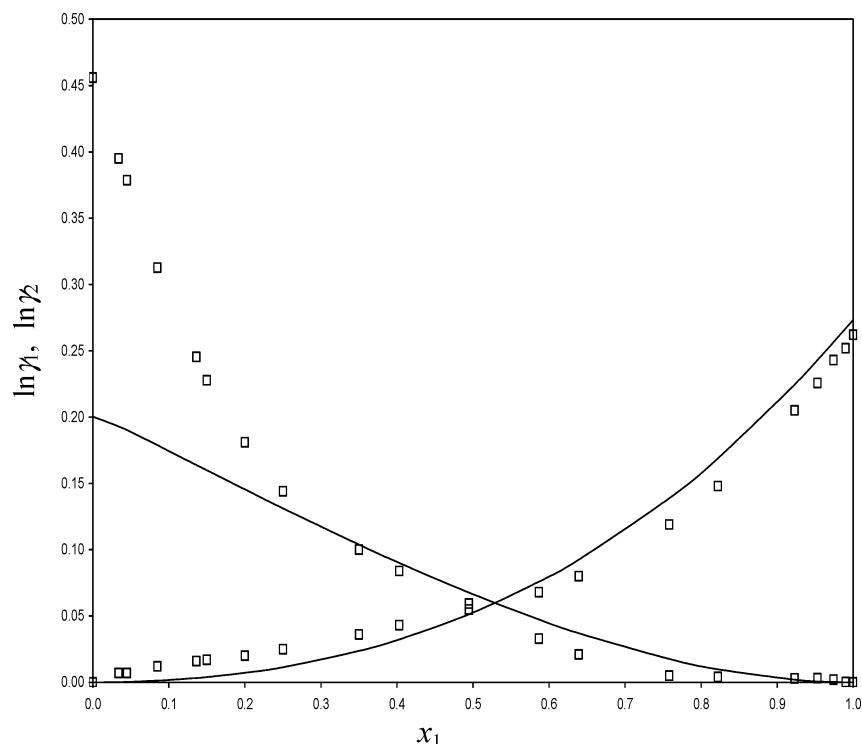


Figure 3. Plot of $\ln \gamma_1$, $\ln \gamma_2$ vs x_1 for the system *p*-xylene (1) + decane (2) at 97.3 kPa: \square , experimental; —, UNIFAC.

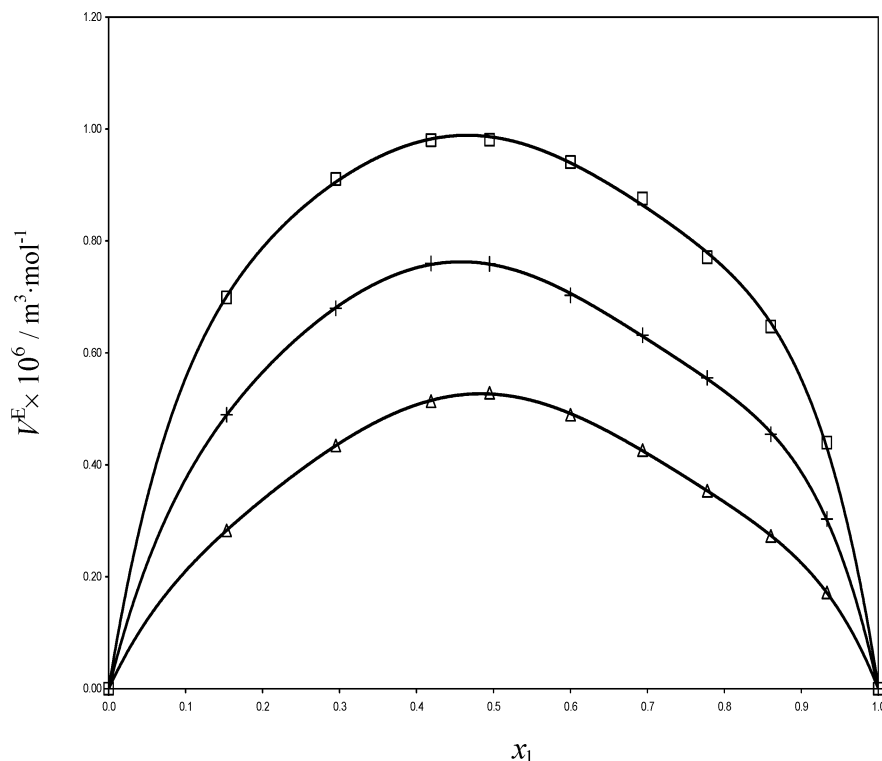


Figure 4. Excess molar volume (V^E) for the system *p*-xylene (1) + decane (2) at \triangle , 303.15 K; +, 308.15 K; and \square , 313.15 K.

The values of mixture density (ρ_m) and speed of sound (u) are used to calculate the isentropic compressibility (K_S) by using the relation:

$$K_S = u^{-2} \rho_m^{-1} \quad (7)$$

The excess isentropic compressibility (K_S^E) was obtained from the relation:

$$K_S^E = K_S - K_S^{\text{id}} \quad (8)$$

where K_S is the experimental compressibility and K_S^{id} is the isentropic compressibility of an ideal mixture of the components. The values of K_S are given in Table 5.

K_S^{id} was determined by using the Kiyohara and Benson¹² equation:

$$K_S^{\text{id}} = \sum \phi_i \left[k_{S,i}^0 + \frac{TV_i^0(\alpha_i^0)^2}{C_{Pi}^0} \right] - \left[\frac{T(\sum x_i V_i^0)(\sum \phi_i \alpha_i^0)^2}{\sum x_i C_{Pi}^0} \right] \quad (9)$$

where ϕ_i is the volume fraction of component i in the

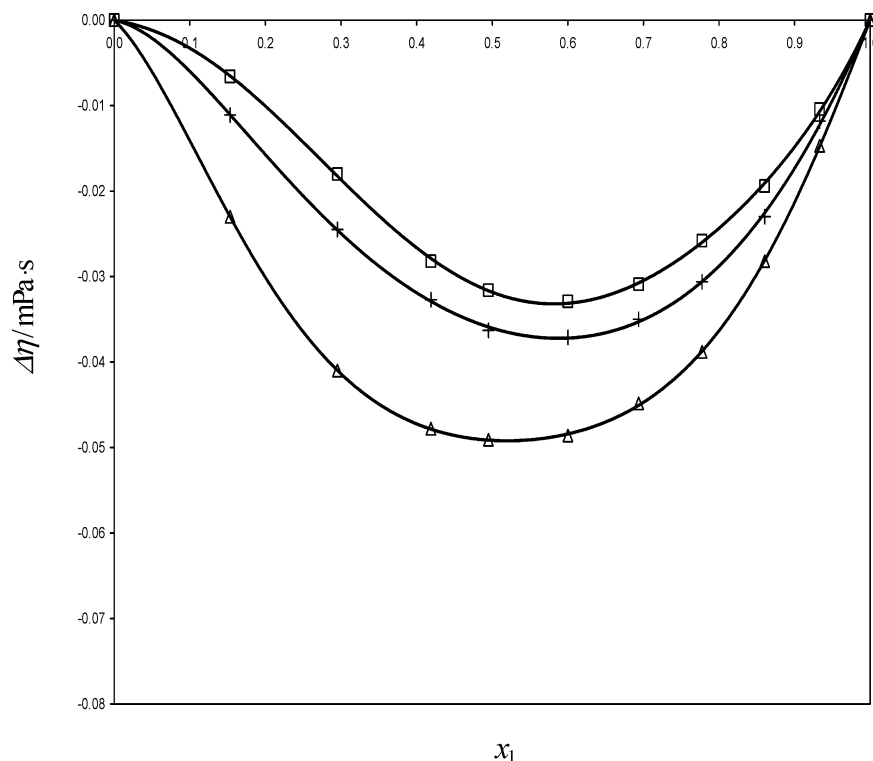


Figure 5. Viscosity deviations ($\Delta\eta$) for the system *p*-xylene (1) + decane (2) at Δ , 303.15 K; +, 308.15 K; and \square , 313.15 K.

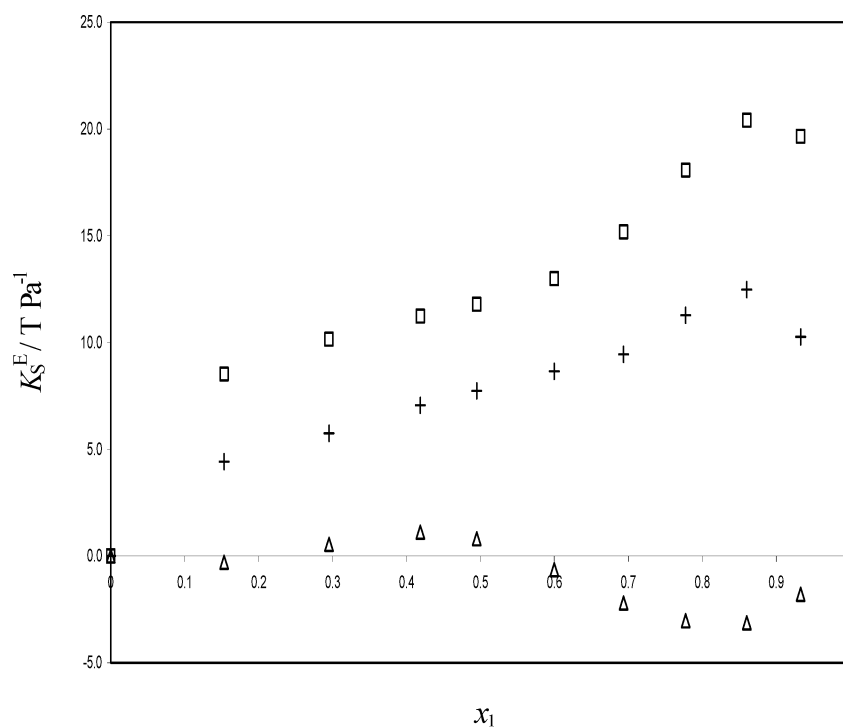


Figure 6. Excess compressibility (K_S^E) for the system *p*-xylene (1) + decane (2) at Δ , 303.15 K; +, 308.15 K; and \square , 313.15 K.

mixture stated in terms of the unmixed components, T is the temperature, and $h_{S,i}^0$, V_i^0 , α_i^0 , and $C_{P,i}^0$ are the isentropic compressibility, molar volume, coefficient of thermal expansion, and molar heat capacity, respectively, for pure component i . The values of α_i^0 were obtained from the density values at two different temperatures. The values of the molar heat capacity were interpolated from the values given in the literature.^{7,13}

The excess volume (V^E), deviations in viscosity ($\Delta\eta$), and excess isentropic compressibility (K_S^E) were fitted to a Redlich–Kister¹⁴ type equation:

$$A = x_1 x_2 \sum_{j=1}^n A_{j-1} (x_1 - x_2)^{(j-1)} \quad (10)$$

where A is the property under consideration, A_{j-1} is the polynomial coefficient, and n is the polynomial degree.

The standard deviation in each case is calculated using

$$\sigma(X) = \left[\frac{\sum (X_{\text{expt}} - X_{\text{calc}})^2}{N - n} \right]^{1/2} \quad (11)$$

where N is the number of data points and n is the number of coefficients. The values of coefficients of the eq 10 as determined by the method of least squares along with the standard deviations at different temperatures for the system are reported in Table 6.

Discussion

The physical constants of the pure components are given in Table 2. Table 3 gives the experimental T , x , y , and activity coefficients data. The x , y and T , x , y plots for p -xylene + decane system at 97.3 kPa are given in Figure 1 and Figure 2, respectively. Figure 3 gives the comparison of experimental activity coefficient data with those calculated using UNIFAC model.¹⁵ As can be seen from Figure 3, the predictability of activity coefficients by UNIFAC is quite satisfactory except for low mole fractions of p -xylene which may be attributed to the unaccountability of $-\text{CH}_2$ molecules attached to the aromatic ring. The data for the system was assessed for thermodynamic consistency by applying the Herington area test, Black test, and Hirata test and were found to be thermodynamically consistent.

The activity coefficient data were fitted into the Margules, Redlich–Kister, and Black¹⁶ correlations. The constants in these correlations and the corresponding average absolute deviations are given in Table 4. The Redlich–Kister and Margules equations gave better fit to the experimental data as compared to the Black equation.

The positive values of V^E (Figure 4) indicate nonspecific interactions between the real species present in the mixture. The values of V^E become more positive with increase in temperature. The negative values of $\Delta\eta$ (Figure 5) are ascribed to the dominance of dispersion forces.

The positive values of K_S^E (Figure 6) indicate that the mixture is more compressible than the corresponding ideal mixture and hence has large volume due to structure disruption effect. With an increase in temperature, the values of K_S^E become more positive except at 303.15 K, which may be due to free volume effect.

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