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Excess Enthalpies for the Ternary Systems 1-Propanol–Acetonitrile–Benzene and 2-Propanol–Acetonitrile–Benzene at 25 °C

Kazuhiro Tamura and Isamu Nagata*
Department of Chemical Engineering, Kanazawa University, Kanazawa 920, Japan

Excess enthalpies for the ternary systems 1-propanol–acetonitrile–benzene and 2-propanol–acetonitrile–benzene and for the constituent binaries 1-propanol–acetonitrile and 2-propanol–acetonitrile were measured at 25 °C by an isothermal dilution calorimeter. The experimental data were correlated by means of the association model of Nagata and Tamura, based upon mole fraction statistics.

Introduction

As part of continuing studies of excess enthalpies for binary and ternary mixtures containing an alcohol and acetonitrile, excess enthalpies of mixtures formed by acetonitrile with methanol and ethanol have been previously reported (*1*, *2*). In this paper, excess enthalpies at 25 °C for the binary systems 1-propanol–acetonitrile and 2-propanol–acetonitrile and for the ternary systems 1-propanol–acetonitrile–benzene and 2-propanol–acetonitrile–benzene are presented. A comparison is made between the experimental results for the ternary systems and those predicted by the association model proposed by Nagata and Tamura (*1–3*). Excess enthalpies at 25 °C for the five binary systems constituting the ternary systems had been previously reported, namely, 1-propanol–acetonitrile (*4–6*); 1-propanol–benzene (*7*); 2-propanol–acetonitrile (*4*, *5*); 2-propanol–benzene (*7*, *8*); and acetonitrile–benzene (*9*).

Experimental Section

All chemical reagents were supplied by Wako Pure Chemical Industries, Ltd. Analytical grade acetonitrile was used without further purification. C.P. benzene was recrystallized three times. C.P. 1-propanol and 2-propanol were fractionally distilled after drying by means of anhydrous copper sulfate. Pure component densities, determined by means of an Anton Paar densimeter (DMA40) at 25 °C, were in good agreement with literature values (*10*). Excess enthalpies were measured at 25 ± 0.01 °C by an isothermal dilution calorimeter. Detailed descriptions of the apparatus and procedure were given by Nagata and Kazuma (*11*). The experimental uncertainties of the excess enthalpies were ±0.5% over most of the mole fraction range.

Results

The excess enthalpies for the binary systems 1-propanol–acetonitrile and 2-propanol–acetonitrile at 25 °C are measured, and compared with published values. The experimental results obtained in this work are directly available from the authors. The values for the system 1-propanol–acetonitrile obtained in this investigation agree with those from Dohnal et al. (*6*) within experimental error and are about 4% lower than those from Coca Prados (*4*) and Mato and Coca (*5*), near the middle mole fraction range. The data for the system 2-propanol–acetonitrile are slightly higher than those reported in previous papers (*4*, *5*) over the region of high concentration of 2-propanol. The experimental data for the binary systems 1-propanol (1)–acetonitrile (2), 2-propanol (1)–acetonitrile (2), and acetonitrile (2)–benzene (3) were fitted by an unweighted least-squares method with a polynomial equation of the form

$$H_{ij}^{E} = x_i x_j \sum_{k=1}^{m} a_{ij,k}(x_i - x_j)^{k-1} \quad (1)$$

Mrazek and Van Ness (*7*) correlated their experimental data for the 1-propanol (1)–benzene (3) and 2-propanol (1)–benzene (3) systems with the following equation.

$$H_{13}^{E} = (x_1 x_3)10^4 / \sum_{k=1}^{m} a_{13,k}(x_1 - x_3)^{k-1} \quad (2)$$

Table I lists the coefficients $a_{ij,k}$ for the five binary systems studied in this investigation, as well as the standard deviations.

The excess enthalpy data for the ternary systems 1-propanol (1)–acetonitrile (2)–benzene (3) and 2-propanol (1)–acetonitrile (2)–benzene (3) at 25 °C are presented in Table II and fitted to an equation of the form

$$H_{123}^{E} = H_{12}^{E} + H_{13}^{E} + H_{23}^{E} + x_1 x_2 x_3 \Delta_{123} \quad (3)$$

where H_{12}^{E} and H_{23}^{E} were calculated from eq 1, and H_{13}^{E} from eq 2, utilizing the coefficients reported in Table I. The term Δ_{123} is expressed by

$$\Delta_{123}/RT = b_0 - b_1 x_1 - b_2 x_2 - b_3 x_1^2 - b_4 x_2^2 - b_5 x_1 x_2 - b_6 x_1^3 - b_7 x_2^3 - b_8 x_1^2 x_2 \quad (4)$$

The values of coefficients and standard deviations to fit eq 3 and 4 to the ternary mixtures are $b_0 = 7.7164$, $b_1 = 40.8507$, $b_2 = -5.4003$, $b_3 = -67.5767$, $b_4 = 34.2032$, $b_5 = -38.3734$, $b_6 = -33.5449$, $b_7 = -27.1734$, and $b_8 = 47.4144$, and $\sigma = 5.0$ J mol^{-1} for the 1-propanol–acetonitrile–benzene system, and $b_0 = 9.6191$, $b_1 = 47.2671$, $b_2 = 3.8793$, $b_3 = -76.7335$, $b_4 = 16.2596$, $b_5 = -45.5172$, $b_6 = 39.0163$, $b_7 = -14.9527$, and $b_8 = 52.7360$, and $\sigma = 5.7$ J mol^{-1} for the

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Table I. Coefficients $a_{ij,k}$ of Eq 1 and 2, and Standard Deviations σ

system (i–j)	$a_{ij,1}$	$a_{ij,2}$	$a_{ij,3}$	$a_{ij,4}$	$a_{ij,5}$	$a_{ij,6}$	σ, J mol^{-1}
1-propanol–acetonitrile	7315.9	−527.4	1226.0	−78.1	282.4		1.8
2-propanol–acetonitrile	8071.2	−212.3	1328.8	−414.4	521.7		5.3
acetonitrile–benzene	1776.3	−629.4	238.1	−37.9			1.6
1-propanol–benzene	2.6287	1.5119	0.2274	0.5056	−0.1927	0.0603	
2-propanol–benzene	1.9470	0.6406	−0.1599	0.2217	−0.1661	0.1637	

Table II. Experimental Excess Enthalpies for Ternary Mixtures at 25 °C

x_2'	x_1	x_2	H^E, J mol^{-1}
1-Propanol (1)–Acetonitrile (2)–Benzene (3)[a]			
0.2507	0.0159	0.2467	441.1
	0.0587	0.2360	758.0
	0.1292	0.2183	1069.3
	0.2409	0.1903	1300.5
	0.3329	0.1672	1357.8
	0.4039	0.1494	1347.7
	0.4584	0.1358	1311.9
	0.5042	0.1243	1265.3
	0.5406	0.1152	1217.4
	0.5846	0.1041	1149.4
	0.6396	0.0903	1046.2
	0.6846	0.0791	948.7
	0.7198	0.0703	865.3
	0.7500	0.0627	788.3
	0.7744	0.0566	722.5
0.5008	0.0164	0.4926	583.7
	0.0523	0.4746	841.5
	0.1165	0.4424	1073.6
	0.2129	0.3942	1460.3
	0.2874	0.3568	1573.3
	0.3413	0.3298	1609.1
	0.3935	0.3037	1614.6
	0.4441	0.2784	1594.8
	0.4814	0.2597	1565.3
	0.4961	0.2523	1550.6
	0.5519	0.2244	1478.1
	0.5984	0.2011	1398.8
	0.6408	0.1799	1311.1
	0.6729	0.1638	1235.9
	0.7012	0.1496	1162.8
	0.7252	0.1376	1095.8
	0.7460	0.1272	1034.5
	0.7682	0.1161	964.7
	0.7978	0.1013	866.2
	0.8219	0.0892	780.6
	0.8429	0.0787	702.7
	0.8584	0.0709	642.7
	0.8714	0.0644	590.7
0.7488	0.0208	0.7332	569.6
	0.0587	0.7048	835.5
	0.1254	0.6549	1176.5
	0.1184	0.6077	1422.1
	0.2482	0.5629	1585.0
	0.3076	0.5185	1688.6
	0.3594	0.4796	1740.9
	0.4077	0.4435	1760.5
	0.4463	0.4146	1758.0
	0.4734	0.3943	1746.7
	0.4949	0.3782	1732.5
	0.5233	0.3570	1706.3
	0.5701	0.3219	1645.5
	0.6157	0.2877	1565.9
	0.6519	0.2606	1488.3
	0.6810	0.2389	1416.8
	0.7043	0.2214	1353.5
	0.7251	0.2058	1292.2
2-Propanol (1)–Acetonitrile (2)–Benzene (3)[b]			
0.2505	0.0208	0.2453	507.2
	0.0565	0.2364	801.0
	0.1117	0.2226	1108.3
	0.1975	0.2011	1397.5
	0.2905	0.1778	1563.1
	0.3691	0.1581	1618.2
	0.4262	0.1438	1618.8
	0.4711	0.1325	1597.5
	0.5165	0.1211	1556.2
	0.5395	0.1154	1528.4
	0.5614	0.1099	1497.1
	0.6111	0.0974	1411.8
	0.6483	0.0881	1334.1
	0.6856	0.0788	1243.6
	0.7153	0.0713	1163.3
	0.7379	0.0657	1097.1
	0.7604	0.0600	1026.8
	0.7751	0.0563	978.3
0.5005	0.0175	0.4918	611.5
	0.0510	0.4750	874.0
	0.0881	0.4564	1111.1
	0.1520	0.4244	1408.5
	0.2243	0.3883	1630.0
	0.2866	0.3571	1750.9
	0.3408	0.3300	1812.1
	0.3848	0.3079	1836.1
	0.4288	0.2859	1839.0
	0.4594	0.2706	1828.5
	0.4897	0.2554	1808.8
	0.5128	0.2439	1786.6
	0.5677	0.2164	1716.2
	0.6110	0.1947	1638.9
	0.6481	0.1762	1558.0
	0.6779	0.1612	1482.6
	0.7044	0.1480	1407.8
	0.7300	0.1351	1329.0
	0.7358	0.1322	1308.9
	0.7698	0.1152	1189.8
	0.7972	0.1015	1084.5
	0.8177	0.0912	1000.0
	0.8333	0.0835	932.3
	0.8486	0.0758	864.4
0.7506	0.0193	0.7360	566.9
	0.0535	0.7104	831.5
	0.0926	0.6810	1089.7
	0.1419	0.6441	1348.8
	0.2054	0.5964	1603.0
	0.2656	0.5512	1771.8
	0.3159	0.5135	1870.3
	0.3645	0.4770	1932.1
	0.4039	0.4474	1960.3
	0.4432	0.4179	1969.4
	0.4649	0.4016	1966.5
	0.4734	0.3953	1963.8
	0.5256	0.3561	1929.7
	0.5759	0.3183	1868.0
	0.6144	0.2894	1800.8
	0.6450	0.2665	1735.8
	0.6733	0.2452	1665.3
	0.6967	0.2276	1600.0
	0.7077	0.2194	1567.1

[a] Obtained by mixing pure 1-propanol with {(x_2')acetonitrile + (1 − x_2')benzene}. [b] Obtained by mixing pure 2-propanol with {(x_2')acetonitrile + (1 − x_2')benzene}.

Table III. Solvation Equilibrium Constants and Enthalpies of Complex Formation

system (A–B)	temp, °C	K_{A_iB}	$K_{A_iB_j}$	$-h_{A_iB}$, kJ mol^{-1}	$-h_{A_iB_j}$, kJ mol^{-1}
1-propanol–acetonitrile	55	40	35	22.0	16.8
2-propanol–acetonitrile	50	45	40	22.0	16.8
1-propanol–benzene	25	3		8.2	
2-propanol–benzene	25	2.8		8.2	
acetonitrile–benzene	45	0.2[a]		5.2	

[a] 1:1 complex formation is assumed.

2-propanol–acetonitrile–benzene system. Figure 1 shows lines of constant values of excess enthalpies on a triangular diagram for the system 1-propanol–acetonitrile–benzene. Figure 2 is the corresponding ternary diagram for the 2-propanol–acetonitrile–benzene system.

Data Analysis

The excess enthalpies for the binary and ternary systems were correlated by means of the model proposed by Nagata

Table IV. Binary Parameters and Absolute Arithmetic Mean Deviations As Obtained from Excess Enthalpy Data Reduction at 25 °C

system (I–J)	no. of data points	C_I, K	C_J, K	D_I	D_J	α_{IJ}	abs arith mean dev, J mol^{-1}	lit.
1-propanol–acetonitrile	22	−222.32	65.49	−0.9056	−0.8345	0.3	8.2	this work
1-propanol–benzene	10	−1135.43	594.67	−3.8897	2.0141	0.3	7.9	*7*
2-propanol–acetonitrile	19	−627.06	−258.28	−2.7189	−1.5730	0.3	8.0	this work
2-propanol–benzene	17	−1097.98	485.01	−3.6946	0.7566	0.3	7.0	*8*
acetonitrile–benzene	15	399.37	−21.88	2.6005	−0.9393	0.3	1.3	*9*

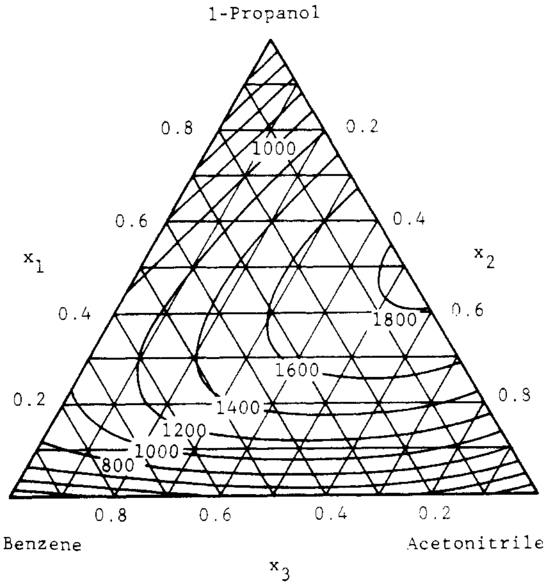

Figure 1. Curves of constant excess enthalpies at 25 °C for the 1-propanol-acetonitrile-benzene system.

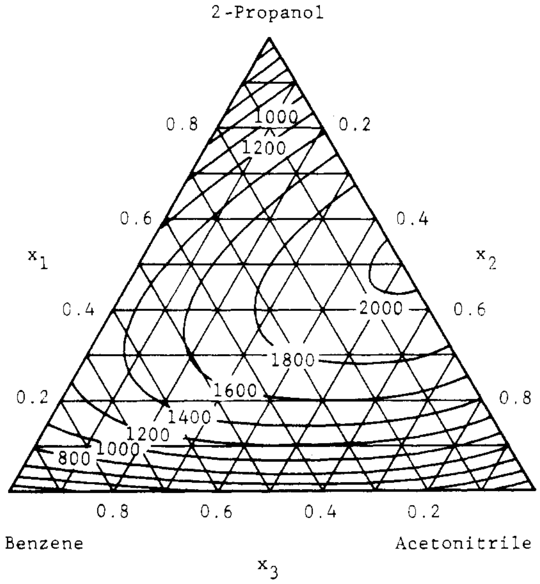

Figure 2. Curves of constant excess enthalpies at 25 °C for the 2-propanol-acetonitrile-benzene system.

and Tamura (*1-3*). This model assumes four equilibrium constants for the association of the propanol, two equilibrium constants for that of acetonitrile, and solvation constants between unlike molecules. Furthermore, the model makes allowance for a nonpolar interaction, given by the NRTL equation (*12*). For the propanol (A)-acetonitrile (B)-benzene (C) system, the association constants of the propanol and acetonitrile are defined in terms of the mole fractions of the chemical species present in the ternary mixtures as follows. For propanol, $K_2 = x_{A_2}/x_{A_1}^2$ for $A_1 + A_1 = A_2$; $K_3 = x_{A_3}/x_{A_2}x_{A_1}$ for $A_1 + A_2 = A_3$; $K = x_{A_{i+1}}/x_{A_i}x_{A_1}$ for $A_1 + A_i = A_{i+1}$ ($i \geq 3$); $K_{cy} = \theta/i$ for A_i(linear) = A_i(cyclic) ($i > 4$), θ being independent of i. For acetonitrile, $K_B' = x_{B_2}(\text{cyclic})/x_{B_1}^2$ for $B_1 + B_1 = B_2$(cyclic); $K_B = x_{B_{i+1}}/x_{B_i}x_{B_1}$ for $B_1 + B_i = B_{i+1}$ ($i \geq 1$). The solvation constants between propanol, acetonitrile, and benzene are as follows: $K_{A_iB} = x_{A_iB}/x_{A_i}x_{B_1}$ for $A_i + B_1 = A_iB$ ($i \geq 1$); $K_{A_iB_j} = x_{A_iB_j}/x_{A_i}x_{B_j}$ for $A_i + B_j = A_iB_j$ ($i \geq 1$, $j > 1$); $K_{A_iC} = x_{A_iC}/x_{A_i}x_{C_1}$ ($i \geq 1$); $K_{BC} = x_{BC}/x_{B_1}x_{C_1}$ for $B_1 + C_1 = BC$.

The excess enthalpies for ternary systems are given by the sum of chemical and physical contributions:

$$H^E = H_{chem}{}^E + H_{phys}{}^E \tag{5}$$

The chemical term of the excess enthalpy, whose derivation is presented in previous papers (*1-3*), is expressed by eq 17 in ref *1*. The monomer mole fraction of the components in the ternary mixtures are obtained by solving the mass balance eq 25-28 in ref *1*, and those for the associated components in pure liquid state are calculated from eq 29 and 30 in ref *1*. Subroutine NOLBR built in a FACOM M-170F computer was used for these purposes. The physical term of the excess enthalpy is given by eq 21 in ref *1*. The coefficients τ_{JI} and G_{JI} are

$$\tau_{JI} = (g_{JI} - g_{II})/T \tag{6}$$

$$G_{JI} = \exp(-\alpha_{JI}\tau_{JI}) \tag{7}$$

where the nonrandomness parameter $\alpha_{JI}(=\alpha_{IJ})$ is set to 0.3. The binary parameters are assumed to be linearly dependent on temperature.

$$g_{JI} - g_{II} = C_I + D_I(T - 273.15) \tag{8}$$

Binary Correlation. The thermodynamic association parameters for the pure associated components are as follows: for acetonitrile (*1-3, 9*), $K_B' = 8.35$ and $K_B = 2.1$ at 45 °C, $h_B' = -8.9$ kJ mol^{-1}, and $h_B = -6.7$ kJ mol^{-1}; for 1-propanol (*3, 13, 14*), $K_2 = 35$, $K_3 = 90$, $K = 40$, and $\theta = 75$ at 25 °C; for 2-propanol (*3, 13, 14*), $K_2 = 35$, $K_3 = 85$, $K = 30$, and $\theta = 70$ at 25 °C, $h_2 = -21.2$ kJ mol^{-1}, and $h_A = -23.5$ kJ mol^{-1} for the propanol (*3, 13, 14*). The values for the solvation constant and enthalpies of complex formation between unlike molecules are presented in Table III (*3*). These enthalpies of the complex formation were assumed to be independent of temperature, and the temperature dependence of the equilibrium constants were calculated by the van't Hoff relation. Table IV presents the binary parameters and absolute arithmetic mean deviations obtained by minimizing the sum of the deviations between the experimental values for the binary systems and the values calculated by the simplex method (*15*).

Ternary Prediction. The excess enthalpies for the ternary systems are presented by use of the association model with the binary parameters obtained solely from the binary correlation. The values of the absolute arithmetic mean deviation between the experimental results and predicted ones are 21.5 J mol^{-1} for the 1-propanol-acetonitrile-benzene system, and 18.4 J mol^{-1} for the 2-propanol-acetonitrile-benzene system.

Glossary

A, B, C,	alcohol, acetonitrile, and benzene
$a_{ij,k}$	coefficient of eq 1 and 2
b_i	coefficient of eq 4
C_I, D_I	constants of eq 8
G_{IJ}	$\exp(-\alpha_{IJ}\tau_{IJ})$
g_{IJ}	binary interaction parameter
H^E	molar excess enthalpy
h_2	enthalpy of hydrogen-bond formation for alcohol dimer
h_A	enthalpy of hydrogen-bond formation for alcohol higher polymer, including cyclic polymer
h_{A_iB}	enthalpy of complex formation between alcohol i-mer and acetonitrile
$h_{A_iB_j}$	enthalpy of complex formation between alcohol i-mer and acetonitrile j-mer ($j > 1$)
h_{A_iC}	enthalpy of complex formation between alcohol i-mer and benzene
h_B'	enthalpy of formation for head-to-head dimerization of acetonitrile
h_B	enthalpy of formation for head-to-tail chain association of acetonitrile
h_{BC}	enthalpy of complex formation between acetonitrile and benzene
K_2	association constant for dimer formation of alcohol

K_3	association constant for open-chain trimer formation of alcohol
K	association constant for open-chain *i*-mer formation of alcohol, $i > 3$
K_{cy}	association constant for cyclization of open *i*-mer as defined by θ/i, $i > 4$
K_{A_iB}	solvation constant for complex formation between alcohol *i*-mer and acetonitrile
$K_{A_iB_j}$	solvation constant for complex formation between alcohol *i*-mer and acetonitrile *j*-mer ($j > 1$)
K_{A_iC}	solvation constant for complex formation between alcohol *i*-mer and benzene
K_B'	association constant for head-to-head dimerization of acetonitrile
K_B	association constant for head-to-tail chain association of acetonitrile
K_{BC}	solvation constant for complex formation between acetonitrile and benzene
R	universal gas constant
T	absolute temperature
x_I	mole fraction of component I

Greek Letters

α_{IJ}	nonrandomness parameter of NRTL equation
Δ_{123}	function defined by eq 4
θ	constant related to K_{cy}
τ_{IJ}	$(g_{IJ} - g_{JJ})/T$

Subscripts

chem	chemical
phys	physical
I, J, *i*, *j*	component

Registry No. 1-Propanol, 71-23-8; 2-propanol, 67-63-0; acetonitrile, 75-05-8; benzene, 71-43-2.

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Vapor-Liquid Equilibrium of Mixtures Formed by Methanol, 2-Propanol, and Chlorobenzene

Isamu Nagata

Department of Chemical Engineering, Kanazawa University, Kanazawa 920, Japan

Isothermal vapor-liquid equilibrium data at 55 °C are presented for the binary system 2-propanol-chlorobenzene and the ternary system methanol-2-propanol-chlorobenzene. The experimental results are well correlated with the UNIQUAC associated-solution theory with binary parameters obtained from the maximum likelihood principle.

Introduction

The number of papers on vapor-liquid equilibrium (VLE) of ternary mixtures containing two alcohols is small. This work reports isothermal VLE at 55 °C for the methanol-2-propanol-chlorobenzene and 2-propanol-chlorobenzene systems. VLE at 55 °C for two of the binary systems of the ternary system have already been published in the literature: methanol-2-propanol (*1*) and methanol-chlorobenzene (*2*).

Experimental Section

All chemicals were purchased from Wako Pure Chemical Industries Ltd. Spectrograde methanol and special grade 2-propanol and chlorobenzene were used as received. Gas chromatographic analysis did not show any appreciable impurities in the compounds used.

A Boublik vapor-liquid recirculation still was used to obtain VLE data for the binary and ternary systems. The apparatus and operational procedure are the same as described previously (*3*). The compositions of liquid samples of the two phases in equilibrium were analyzed by combining the use of a Shimadzu GC-7A gas chromatograph and an electronic integrator (Shimadzu Chromatopac C-E1B). The measured quantities were considered to have the following experimental errors: 0.002 for mole fraction, 21.3 Pa for pressure, and 0.05 K for temperature.

Table I. Properties Used in Data Analysis

1	methanol (1)-2-propanol (2)	methanol (1)-chlorobenzene (3)	2-propanol (2)-chlorobenzene (3)
P_i^s, kPa	68.301	68.301	30.597
P_j^s, kPa	30.597	7.159	7.159
v_i^L, cm^3/mol	41	41	81
v_j^L, cm^3/mol	81	104	104
B_{ii}, cm^3/mol	-1437	-1437	-1510
B_{jj}, cm^3/mol	-1510	-2248	-2248
B_{ij}, cm^3/mol	-1506	-662	-1112
r_i	1.15	1.15	2.23
r_j	2.23	3.06	3.06
q_i	1.12	1.12	1.98
q_j	1.98	2.45	2.45

Experimental Results and Analysis of Results

Table I lists properties used in experimental data analysis. P^s is the vapor pressure of pure components, v^L is the pure-liquid molar volume calculated from the modified Rackett