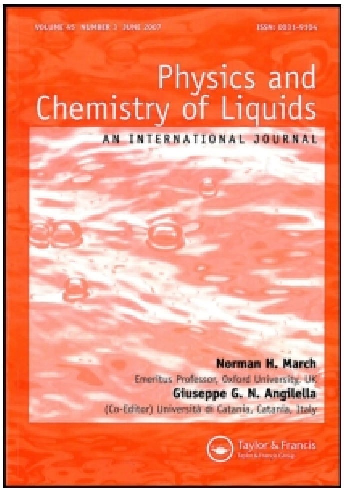

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Excess Enthalpy Data in some Binary Systems Containing Non-Electrolyte Solutions and their Correlations at 298.15 K - Part II

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EXCESS ENTHALPY DATA IN SOME BINARY SYSTEMS CONTAINING NON-ELECTROLYTE SOLUTIONS AND THEIR CORRELATIONS AT 298.15 K – PART II

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Excess enthalpy data measured at 298.15 K were collected from the literature. The data contains 48 binary systems (812 data points). The experimental data were correlated by the Redlich–Kister equation. In this research work the following thermodynamics models were also tested, Wilson, NRTL models and their parameters were calculated. A modified NRTL with composition-dependent (α_{12}) parameter were also proposed. The correlation of excess enthalpy data in the systems using this model provides good results.

Keywords: Excess enthalpy; Redlich–Kister; Wilson model; NRTL model

1. INTRODUCTION

The present work forms part of a program involved in the calculating of the excess enthalpy for some binary liquid mixtures [1–4]. The results can be applied for development of heat exchange, heat transport and heat storage equipments. For non-ideal solutions the magnitude of their excess enthalpies can provide information needed to test existing theories of solutions.

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In this paper we collected some other literature measurements of excess enthalpy at 298.15 K [5] that were correlated by the Redlich–Kister equation. We also test the Wilson and NRTL models for excess enthalpy of mixing H_m^E, to investigate which model gives the best correlation of excess enthalpy and to calculate their parameters. Finally for nine systems, namely propylene corbonate + methyl acetate and ethyl octanoate + oxane, oxolane and 1,3-dioxaolane; diethyl carbonate + oxane and oxolane; propylene carbonate + oxolane and 1,4-dioxan; 1,2,4-Trimethyl benzene + oxane and oxolane, the deviations between calculated and experimental H_m^E are large based on the Wilson and NRTL equations. Therefore we have used a modified NRTL model with composition-dependent (α_{12}) parameter.

2. DATA BASE OF EXCESS ENTHALPY

Tables I–IV summarize the references of excess enthalpy data collected from the literature. The data base is divided into four parts according to the nature of the components.

2.1. Propylene carbonate + Alkanoate systems

This part of data sets contains 413 data points of 27 binary mixtures. The binary systems of part one are listed in Table I.

2.2. Cyclic ethers + Hydrocarbons systems

The second part of the data base contains 100 data points of 5 binary mixtures. The binary systems of part two are listed in Table II.

3. ANALYSIS OF THE DATA BASE

The following cases are found when analyzing the excess enthalpy data:

1. The excess enthalpy-composition plot changes sign and goes from positive to negative values [6,7].

TABLE I Excess enthalpy data of propylene carbonate + alkanoate systems at 298.15 K

Systems	*Data points*	*System no.*
propylene carbonate[a] + methyl acetate	17	1–1
ethyl acetate	17	1–2
vinyl acetate	17	1–3
propyl acetate	17	1–4
butyl acetate	17	1–5
pentyl acetate	17	1–6
hexyl acetate	17	1–7
methyl propanoate	16	1–8
ethyl propanoate	16	1–9
propyl propanoate	16	1–10
butyl propanoate	16	1–11
methyl butyrate	16	1–12
ethyl butyrate	16	1–13
propyl butyrate	16	1–14
butyl butyrate	16	1–15
ethyl pentanoate	16	1–16
ethyl hexanoate	16	1–17
ethyl heptanoate	16	1–18
ethyl octanoate	16	1–19
tert-butyl acetate	17	1–20
ethyl isobutyrate	16	1–21
isobutyl acetate	16	1–22
methyl pentanoate	16	1–23

[a] Ref. [10].

2. The excess enthalpies show large negative or positive values greater than 550 J mol^{-1} [8–11].
3. The excess enthalpies show moderate negative or positive values less than 550 J mol^{-1} [6–9].
4. The excess enthalpy-composition plot is symmetric or asymmetric [6–11].

4. DATA REDUCTION BY REDLICH–KISTER EQUATION

The Redlich–Kister equation (R–K) [12] is a flexible function commonly used for correlation binary excess enthalpy data. It is an empirical power expansion

$$H_m^E/(\mathrm{J\,mol^{-1}}) = x_1x_2\sum_{K=0}^{N-1} A_K(x_1 - x_2)^K. \qquad (1)$$

TABLE II Excess enthalpy data of cyclic ethers + hydrocarbons systems at 298.15 K

Systems	*Data points*	*System no.*
dimethy carbonate[a] + Oxane	17	2–1
oxolane	17	2–2
1,4-dioxane	17	2–3
1,3-dioxolane	17	2–4
diethyl carbonate[a] + oxane	17	2–5
oxolane	17	2–6
1,4-dioxane	17	2–7
1,3-dioxolane	17	2–8
propylene carbonate[b] + oxane	17	2–9
oxolane	17	2–10
1,4-dioxane	18	2–11
1,3-dioxolane	17	2–12
1,2,4-trimethylbenzene[c] + oxane	14	2–13
1,4-dioxane	15	2–14
oxolane	15	2–15
methanol[d] + tetrahydrofuran	15	2–16
tetrahydropyran	15	2–17
1,4-dioxane	18	2–18
ethanol[d] + tetrahydrofuran	16	2–19
tetrahydropyran	18	2–20
1,4-dioxane	21	2–21
propan-1-ol[d] + tetrahydrofuran	18	2–22
tetrahydropyran	17	2–23
1,4-dioxane	19	2–24
propan-2-ol[d] + tetrahydofuran	16	2–25
tetrahydropyran	18	2–26
1,4-dioxane	19	2–27
1,4-dioxane[e] + heptane	17	2–28
octane	17	2–29
nonane	16	2–30
decane	17	2–31
dodecane	17	2–32
tetradecane	17	2–33
hexadecane	17	2–34
1,3-dionolane[f] + heptane	17	2–35
octane	17	2–36
nonane	17	2–37
decane	17	2–38
dodecane	17	2–39
tetradecane	9	2–40

[a]Ref. [7]; [b]Ref. [6]; [c]Ref. [8]; [d]Ref. [9]; [e]Ref. [10]; [f]Ref. [11].

Where A_K is an adjustable parameter, x_i ($i = 1$ or 2) is the mole fraction of component i, and N is the number of adjustable parameters. All data sets of the excess enthalpy were correlated by Redlich–Kister equation, and the results are given in the literature [6–11].

5. CORRELATION BY WILSON AND NRTL MODELS

5.1. Wilson Equation [13]

In the development of the equation, Wilson conceived that the interaction between molecules depends primarily on "local concentrations", which he expressed as volume fractions. These concentrations are defined in probabilistic terms, the Boltzman distribution of energies. In a binary mixture that consists of molecules of types 1 and 2, in the vicinity of a molecule type 1 the probability of finding other molecules of the same type is P_{11} and the probability of finding the other type is P_{12}. Accordingly the ratio of the two probabilities are:

$$\frac{P_{21}}{P_{11}}=\frac{x_2\exp(-a_{21}/RT)}{x_1\exp(-a_{11}/RT)}. \tag{2}$$

where a_{21} and a_{11} representing the different interaction energies.

By means of this relation the following expression for H_m^E is obtained

$$\frac{H^E}{x_1x_2}=\frac{\lambda_{12}\Lambda_{12}}{x_1+\Lambda_{12}x_2}+\frac{\lambda_{21}\Lambda_{21}}{\Lambda_{21}x_1+x_2}, \tag{3}$$

where $\Lambda_{12}=(V_2/V_1)\exp(-(\lambda_{12}/RT))$, $\Lambda_{21}=(V_1/V_2)\exp(-(\lambda_{21}/RT))$, $\lambda_{12}=a_{21}-a_{11}$, $\lambda_{21}=a_{12}-a_{22}$ and $a_{12}=a_{21}$ where V_i is molar volume.

5.2. NRTL Equation [14]

To take into account non-randomness in this model, the following relation between the local mole fraction x_{11} of molecules 1 and the local mole fraction x_{21} of molecules 2 which are in the immediate neighborhood of molecules 1 is given by

$$\frac{x_{21}}{x_{11}}=\frac{x_2\exp(-\alpha_{21}g_{21}/RT)}{x_1\exp(-\alpha_{21}g_{11}/RT)}, \tag{4}$$

where g_{21} and g_{11} indicate the different interaction energies. The non-randomness parameter α_{21} is an empirical constant.

The following expression for H_m^E is obtained with this relation

$$H^E(x) = -x_1x_2\left[\frac{Z_1N_1\alpha_{12}\Delta g_{21} - \Delta g_{21}G_{21}N_1 - Z_1x_2G_{21}\alpha_{12}\Delta g_{21}}{N_1^2} + \frac{Z_2N_2\alpha_{12}\Delta g_{12} - \Delta g_{12}G_{12}N_2 - Z_2x_1G_{12}\alpha_{12}\Delta g_{12}}{N_2^2}\right], \tag{5}$$

where $\Delta g_{21} = g_{21} - g_{11}$; $\Delta g_{12} = g_{12} - g_{12}$; $G_{21} = \exp(-\alpha_{12}\Delta g_{21}/RT)$; $G_{12} = \exp(-\alpha_{12}\Delta g_{12}/RT)$; $Z_1 = (\Delta g_{21}/RT)G_{21}$; $Z_2 = (\Delta g_{12}/RT)G_{12}$; $N_1 = x_1 + x_2G_{21}$; $N_2 = x_2 + x_1G_{12}$; Δg_{21} and Δg_{12} are the two adjustable parameters remaining, because the randomness parameters α_{21} were taken to be constant for each of the binary mixtures studied here.

6. MODIFIED NRTL EQUATION

We modified the NRTL equation by expressing α_{12} as a function of composition. The composition-dependent α_{12} is similar to the Redlich–Kister expansion

$$\alpha_{12} = \sum_{i=0}^{N} B_i(1 - 2x)^{i-1}, \tag{6}$$

where the B_i are adjustable parameters and x is the mole fraction of the first component. For calculating α_{12} as a function of x based on Eq. (6) the same Δg_{21}, Δg_{12} that were calculated for the NRTL equation were applied, we used Tables III and IV, and experimental excess enthalpy [7–12]. For each mole fraction, α_{12} was found for minimum deviation between experimental and calculated excess enthalpy. Finally the parameters of Eq. (6) were obtained by a least squares fit.

Using Eq. (5), where α_{12} are replaced with the ones given by Eq. (6), we can calculate the excess enthalpy by the modified NRTL equation.

7. RESULTS AND DISCUSSION

The results of data correlation by the Wilson and NRTL models for 48 selected systems are given in Tables III and IV for each systems and the absolute deviation (AD) and relative deviation (RD) based on following equations were calculated

$$\mathrm{AD} = \frac{\sum \left| H^E_{\mathrm{exp}_i} - H^E_{\mathrm{cal}_i} \right|}{N} \tag{7}$$

$$\mathrm{RD}\% = \frac{\mathrm{AD} \times 100}{H^E_{(\mathrm{exp})_{\max}}} \tag{8}$$

where N is the number of data point and $H^E_{(\mathrm{exp})_{\max}}$ is the maximum experimental absolute excess enthalpy in each system. For eleven systems,

TABLE III Adjustable parameters, Δg_{ij}, λ_{ij}, α_{12}, absolute deviation (AD) and relative deviation (RD) based on NRTL and Wilson models for calculating excess molar enthalpies for propylene carbonate + alkanoates at 298.15 K

System no.	*NRTL*					*Wilson*			
	Δg_{12}	Δg_{12}	α_{12}	*AD*	*RD%*	λ_{12}	λ_{21}	*AD*	*RD%*
1–1	−351.4	565.0	0.50	2.9	11	4484	−253.1	2.7	10.3
1–2	−498.6	589.8	1.60	3.2	1.8	615.8	160.7	3.5	2.0
1–3	1040.2	398.9	0.25	1.8	0.56	903.6	589.8	1.9	0.59
1–4	391.5	1401.7	1.00	4.0	1.6	1913.2	−274.4	4.4	1.7
1–5	1766.0	1407.3	0.60	4.1	0.84	1472.0	1070.1	7.8	1.6
1–6	2535.2	1890.1	0.50	3.3	0.53	1929.7	1853.1	6.2	0.99
1–7	3473.9	2108.0	0.40	16.8	2.3	2764.2	3798.8	12.4	1.7
1–8	234.5	626.4	0.70	3.1	1.7	755.0	66.5	3.0	1.6
1–9	1000.0	1000.0	0.60	5.6	1.5	1292.4	499.4	5.8	1.5
1–10	1572.2	1582.1	0.50	7.5	1.4	1811.2	1079.1	7.3	1.3
1–11	2304.0	1766.1	0.40	5.7	0.83	2297.6	2199.6	8.4	1.2
1–12	928.8	1037.5	0.80	3.9	1.2	1186.3	361.1	4.9	1.5
1–13	1567.0	1592.0	0.50	0.8	1.3	1823.0	1091.6	6.9	1.3
1–14	2276.5	1890.2	0.40	5.9	0.85	2441.6	2152.4	7.0	1.0
1–15	3712.6	2935.0	0.40	7.3	0.85	3306.5	3324.8	34.6	4.2
1–16	2370.0	1864.0	0.40	7.6	1.1	2424.0	2234.1	4.4	0.63
1–17	3711.9	2909.7	0.40	4.4	0.54	3292.6	3344.1	32.8	4.0
1–18	3777.8	2187.4	0.30	25.9	2.9	3520.3	3393.4	89.8	10
1–19	4325.6	2851.7	0.30	44.6	4.7	3869.4	3269.3	131.9	13.8
1–20	1729.1	1541.2	0.40	2.1	0.35	2029.0	1379.4	2.5	0.40
1–21	1620.3	1041.2	0.30	5.9	1.1	1655.5	1298.8	7.0	1.2
1–22	1672.0	1508.0	0.60	3.3	0.66	1602.2	931.5	7.3	1.5
1–23	1529.6	1278.1	0.50	2.6	0.51	1572.2	1003.9	3.1	0.62

TABLE IV Adjustable parameters, Δg_{ij}, λ_{ij}, α_{12}, absolute deviation (AD) and relative deviation (RD) based on NRTL and Wilson models for calculating excess molar enthalpies for cyclicethers + Hydrocarbons systems at 298.15 K

System no.	*NRTL*					*Wilson*			
	Δg_{12}	Δg_{12}	α_{12}	*AD*	*RD%*	λ_{12}	λ_{21}	*AD*	*RD%*
2–1	236.4	1982.9	0.10	26.1	4.7	1753.1	1153.2	28.4	5.2
2–2	−852.1	2167.2	0.10	20.7	6.7	915.1	446.5	329.9	19.4
2–3	−110.4	777.5	0.10	2.1	1.3	443.2	256.6	2.1	1.3
2–4	1863.8	−1533.0	0.10	0.92	3.5	−653.2	835.5	0.84	3.2
2–5	−2205.9	3815.7	0.10	9.8	4.2	1211.3	41.1	10.5	4.5
2–6	−2491.8	3490.2	0.10	7.9	7.5	877	−165.4	8.9	8.4
2–7	−1030.3	2426.1	0.10	2.9	1.0	659.7	624.1	3.6	1.3
2–8	1444.3	2428.2	0.10	2.9	1.7	138.1	484.6	3.4	2.0
2–9	−177.6	2304.8	0.10	9.3	1.9	1644.2	924.7	10.8	2.1
2–10	434.7	328.4	0.10	13.8	6.6	305.1	555.0	12.8	6.1
2–11	−317.0	2201.5	1.3	3.4	4.4	3758.9	−1254.4	2.7	3.5
2–12	−1982.4	1927.7	0.20	0.9	0.54	1962.4	−1287.7	1.9	1.2
2–13	1127.4	−1777.1	0.40	3.2	14	−1281.1	1763.0	5.5	24.3
2–14	−851.7	3144.6	0.10	10.5	2.2	1195.0	1213.4	13.5	2.8
2–15	−2085.0	−72.2	−0.65	9.1	3.2	4959.1	−1248.5	56.1	19.9
2–16	−2401.2	6274.2	0.10	7.4	1.4	3694.0	−312.3	8.6	1.6
2–17	1436.5	3733.7	0.40	6.0	0.85	4406.7	349.7	5.5	0.78
2–18	1601.3	4067.1	0.15	12.2	1.1	4261.2	3055.3	200.0	17.7
2–19	−14.3	4284.0	0.15	6.5	0.81	4351.9	2198.6	54.8	6.8
2–20	2176.9	4687.9	0.35	14.1	1.7	4145.5	2755.2	94.1	11.0
2–21	4662.2	5352.3	0.20	8.6	0.56	3677.7	3318.1	500.4	32.7
2–22	251.2	4213.3	0.10	8.2	0.86	3603.2	3242.3	159.0	16.7
2–23	−407.4	5073.2	0.10	7.9	0.85	3957.3	2751.1	101.6	10.9
2–24	4185.1	4848.4	0.10	24.2	1.3	3678.8	3490.0	706.5	38.1
2–25	3096.2	4070.3	0.25	19.5	1.7	3597.2	3340.0	260.9	23.3
2–26	2560.9	4648.8	0.25	6.9	0.61	3666.5	3156.7	268.7	23.9
2–27	7345.2	6490.8	0.15	13.2	0.64	3477.5	3441.5	846.4	41.2
2–28	5699.5	4773.1	0.15	13.6	0.76	3662.1	3027.9	792.8	44.4
2–29	6468.0	4861.0	0.15	17.5	0.94	3915.3	3615.7	839.1	45.1
2–30	7422.0	5257.3	0.15	14.1	0.72	3826.0	3228.4	899.8	45.9
2–31	8392.2	5677.6	0.15	16.5	0.81	3639.6	3354.7	978.9	48.0
2–32	8604.4	3520.3	0.10	27.1	1.3	3955.2	2945.5	1005.8	46.4
2–33	9204.8	3690.0	0.10	33.4	1.5	4272.7	2874.5	1049.2	46.3
2–34	9798.8	4092.9	0.10	38.0	1.6	4242.1	2925.1	1094.9	46.3
2–35	6641.1	4519.2	0.15	14.3	0.74	3911.4	3222.6	889.5	45.8
2–36	7503.2	6012.7	0.15	14.1	0.69	3946.6	3068.8	937.6	46.2
2–37	8531.5	6323.4	0.15	20.0	0.96	4062.9	3020.2	968.6	46.4
2–38	9560.1	6948.9	0.15	40.3	1.9	4136.3	3004.9	998.3	46.3
2–39	9301.6	5845.9	0.13	31.9	1.4	4278.4	3080.5	1046.1	46.3
2–40	9257.0	4180.6	0.10	70.6	2.9	4185.1	3069.5	1015.9	43.3

deviations are relatively large (greater than three), this includes propylene carbonate + methyl acetate and ethyl octanoate [14], dimethyl carbonate + oxane, oxolane and 1,3-dioxolane [8], diethyl carbonate + oxane and oxolane [7], propylene carbonate + oxolane and 1,4-diox-

TABLE V Parameters of modified NRTL equation for binary mixtures at 298.15 K

System	Δg_{12}	Δg_{21}	B_1	B_2	B_3	B_4	B_5	B_6	*AD*	*RD%*
propelene carbonate + methyl	−351.4	565.0	0.611856	0.39612	−0.1455	−0.866033	−0.58017		0.69	2.7
acetate, ethyl octanoate	4325.6	2851.7	0.317682	−0.142481	−0.0224357	0.305129	−0.17317		7.6	0.80
dimethyl carbonate + oxane,	236.4	1982.9	0.0244982	0.0341383	−0.567384	−0.576092	2.11991	1.62271	4.0	0.73
1,3-dioxolane	−852.1	2167.2	0.0402001	−0.0710846	−0.0501175	0.241628	0.640889		2.5	0.82
	1683.8	−1533.0	0.976834	−0.003479	0.00889777	0.0078488			0.35	1.3
diethyl carbonate + oxane,	−2205.9	3815.7	0.0914212	−0.0113275	−0.00228388	0.209929	0.702704		1.4	0.62
oxolane	−2421.8	3490.2	0.0919186	−0.00141778	0.00825923	−0.0290868	0.0442676	0.0410091	0.85	0.81
propylene carbonate +	433.7	328.4	−0.430866	0.184238	−3.23443	4.20756	11.6525	−9.43081	31.1	1.5
oxolane, 1,4-dioxane	−317.0	2201.5	1.25184	−0.029638	0.288955	0.38136	−0.363064	−0.538934	0.98	1.3
1,2,4-trimethyl benzene +	1127.4	−1771.1	−0.411807	−0.0339376	0.0407932	0.0630603	0.0260217		0.16	0.70
oxane, oxolane	−2085.0	−42.2	−0.685814	−0.068588	0.00924068	0.750199	0.377618	−1.07661	2.9	1.0

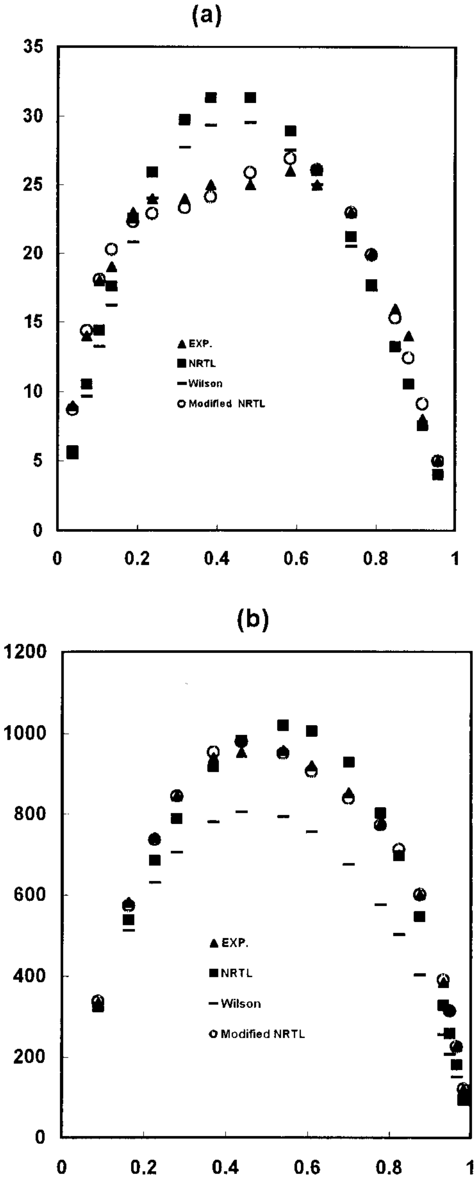

FIGURE 1 Comparison between calculated [Wilson (Eq. (3)), NRTL (Eq. (5)) and modified NRTL (Eqs. (5) and (6)) using coefficients from Table V] and experimental H_m^E (J mol^{-1}) values (Y) for (a) methyl acetate, (b) ethyl octanoate + (x) mole fraction of propylene carbonate at 298.15 K.

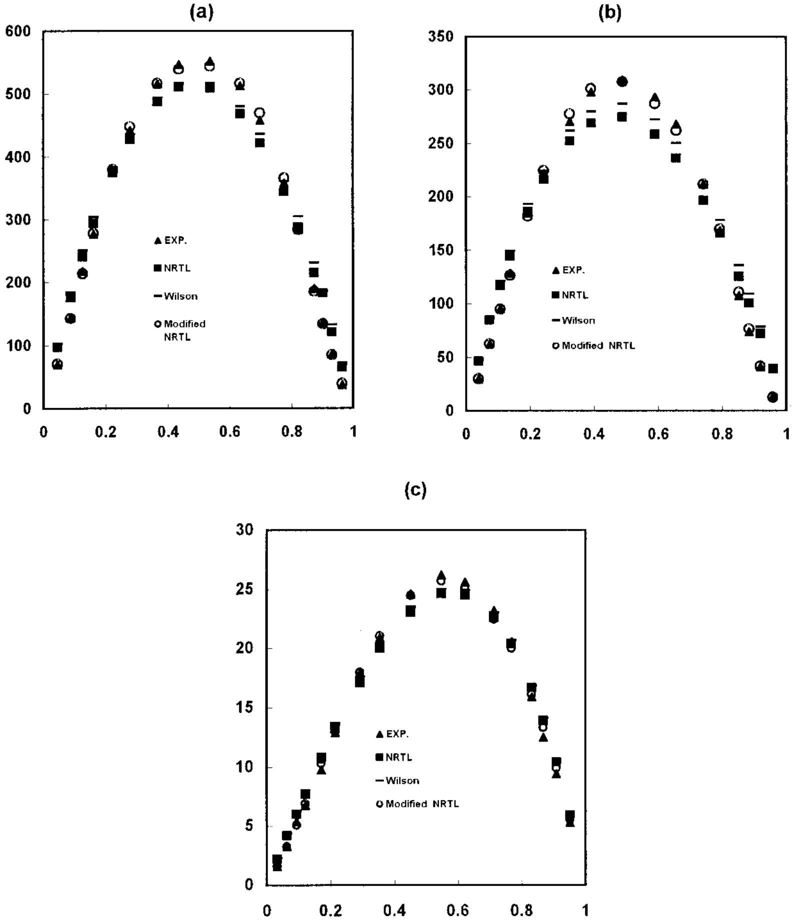

FIGURE 2 Comparison between calculated [Wilson (Eq. (3)), NRTL (Eq. (5)) and modified NRTL (Eqs. (5) and (6)) using coefficients from Table V] and experimental H_m^E (J mol^{-1}) values (Y) for (a) oxane, (b) oxolane, (c) 1,3,-dioxolane + (x) mole fraction of dimethyl carbonate at 298.15 K.

ane [8], 1,2,4-trimethylbenzene + oxane and oxolane [9] were found. The modified NRTL model was applied and the results are summarized in Table V. Figures 1–5 show comparison of the experimental and theoretical H_m^E values as a function of mole fraction derived from the Wilson, NRTL and modified NRTL equations. Figure 6 shows the variation of the relative deviation by different models for

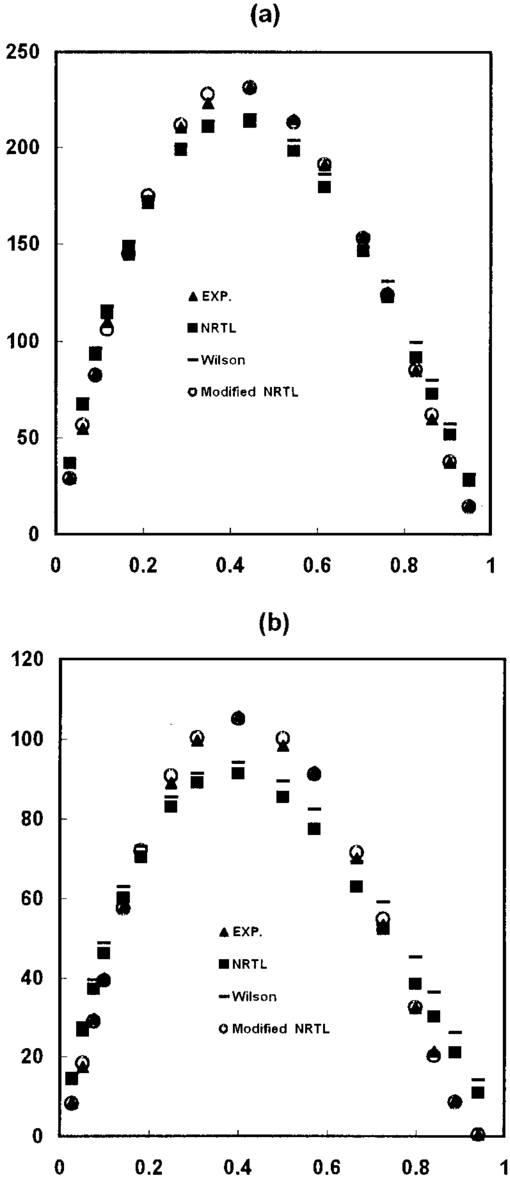

FIGURE 3 Comparison between calculated [Wilson (Eq. (3)), NRTL (Eq. (5)) and modified NRTL (Eqs. (5) and (6)) using coefficients from Table V] and experimental H_m^E ($\mathrm{J\,mol^{-1}}$) values (Y) for (a) oxane, (b) oxolane + (x) mole fraction of diethyl carbonate at 298.15 K.

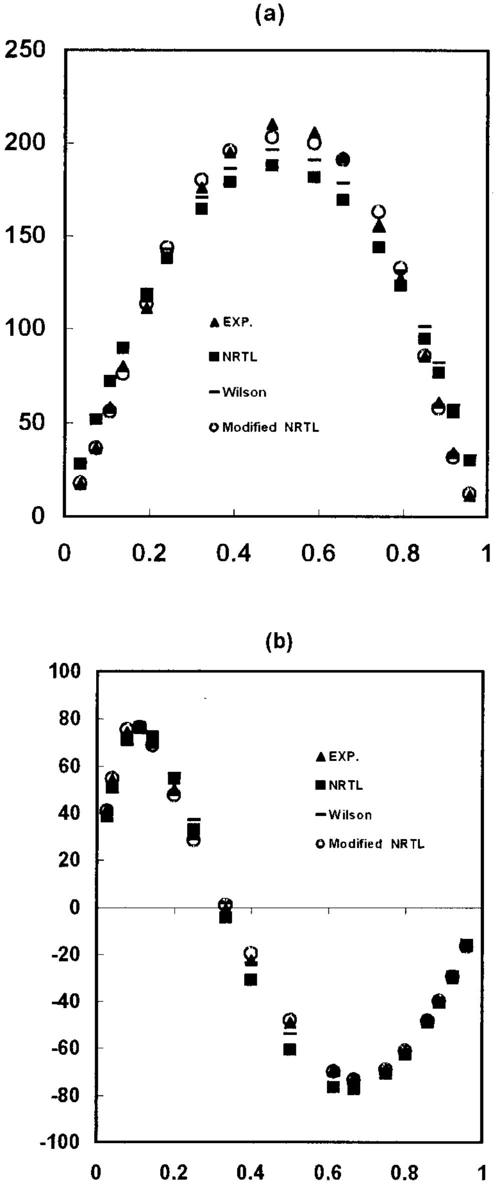

FIGURE 4 Comparison between calculated [Wilson (Eq. (3)), NRTL (Eq. (5)) and modified NRTL (Eqs. (5) and (6)) using coefficients from Table V] and experimental H_m^E (J mol^{-1}) values (Y) for (a) oxolane, (b) 1,4-dioxane + (x) mole fraction of propylene carbonate at 298.15 K.

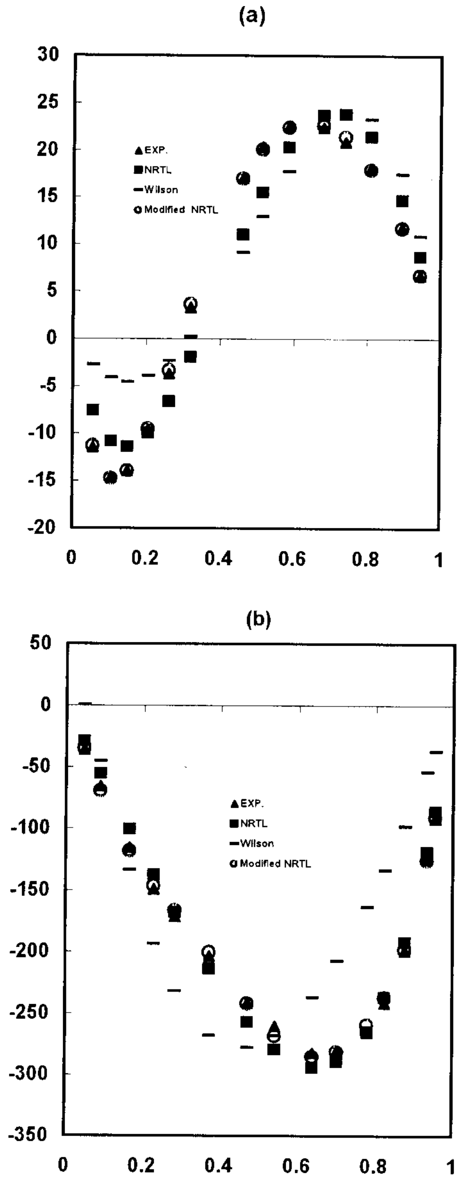

FIGURE 5 Comparison between calculated [Wilson (Eq. (3)), NRTL (Eq. (5)) and modified NRTL (Eqs. (5) and (6)) using coefficients from Table V] and experimental H_m^E (J mol^{-1}) values (Y) for (a) oxolane, (b) oxolane + (x) mole fraction of 1,2,4-trimethylbenzene at 298.15 K.

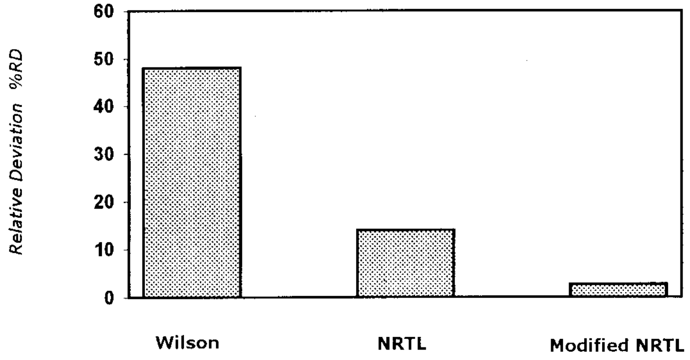

FIGURE 6 Overall deviation by different models.

all systems. The maximum number of adjustable parameters used in the Redlich–Kister equation is 6 for which the best results were then obtained. On the other hand, fewer parameters are necessary for correlation with the Wilson and NRTL models. For modified NRTL models, the relative deviation is smaller than Wilson and NRTL and comparable with Redlich–Kister equations.

In cases when the excess enthalpies are not large and H_m^E–x curves are symmetric, all models tested are able to fit the experimental data with good accuracy. Among them the NRTL equation provides the best fit for such systems. However, when the excess enthalpy curve is asymmetric, the modified NRTL model is an improvement.

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