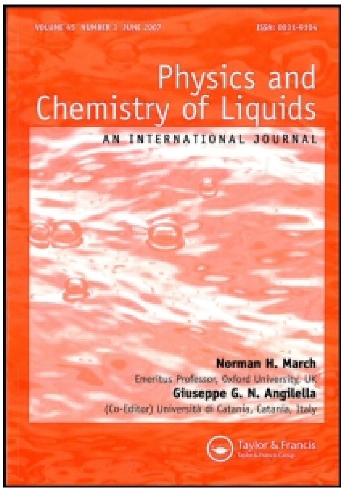

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Excess Enthalpy Data in Some Binary Systems Containing Non Electrolyte Solution and Their Correlations at 298.15K

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EXCESS ENTHALPY DATA IN SOME BINARY SYSTEMS CONTAINING NON ELECTROLYTE SOLUTION AND THEIR CORRELATIONS AT 298.15 K

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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 Redlich-Kister equation. In this work the following thermodynamics models were also tested. Wilson, NRTL models: and their parameters were calculated. A modified NRTL with a composition-dependent (α_{12}) parameter was also proposed. The correlation of excess enthalpy data in the systems using this model provides good results.

Keywords: Excess enthalpy; Redlich-Kister; Wilson; NRTL

1. INTRODUCTION

The present work forms part of a program involved in the calculating of the excess enthalpy for some binary liquid mixtures [1–3]. The results can 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 literature measurements of excess enthalpy at 298.15 K that were correlated by the Redlich-Kister equation, we also tested Wilson and NRTL models for excess enthalpy of mixing H_m^E, to investigate which model gives the best correlation of excess enthalpy and calculate their parameters. Finally for nine systems, namely trichloroethylene + alkan-1-ols and water + alkanolamines, that deviations are large based on Wilson and NRTL equation, we used modified NRTL model with composition-dependent (α_{12}) parameter and calculated their parameters.

2. DATA BASE OF EXCESS ENTHALPY

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

TABLE I Excess enthalpy data of chlorohydrocarbon + hydrocarbon systems at 298.15 K

Systems	*Data points*	*System no.*
tetrachloroethylene[a] +		
propan-1-ol	17	1–1
butan-1-ol	18	1–2
pentan-1-ol	16	1–3
hexan-1-ol	17	1–4
heptan-1-ol	16	1–5
octan-1-ol	16	1–6
trichloroethylene[b] +		
propan-1-ol	17	1–7
butan-1-ol	19	1–8
pentan-1-ol	17	1–9
hexan-1-ol	17	1–10
heptan-1-ol	17	1–11
octan-1-ol	18	1–12
1,1,2,2-tetrachloroethane[c] +		
acetone	9	1–13
dibuthylether	11	1–14
acetonitrile	8	1–15
dimethysolfoxide	10	1–16
2-propyn-1-ol[d] +		
1,2-dichloroethane	14	1–17
1,1,1-trichloroethane	15	1–18
1,1,2,2-tetrachloroethane	15	1–19
trichloroethylene	15	1–20

TABLE I (Continued)

Systems	*Data points*	*System no.*
2-butanone[e] +		
1-chlorobutane	16	1-21
1-chloropentane	16	1-22
1-chlorohexane	16	1-23
1-chloroctane	16	1-24
2-pentanone[e] +		
1-chlorobutane	14	1-25
1-chloropentane	18	1-26
1-chlorohexane	15	1-27

[a] Reference [5].
[b] Reference [4].
[c] Reference [6].
[d] Reference [7].
[e] Reference [10].

TABLE II Excess enthalpy data of ether + hydrocarbon systems at 298.15 K

Systems	*Data points*	*System no.*
methy 1,1-dimethylpropyl ether[a] +		
2-methylpentane	19	2-1
3-methylpentane	19	2-2
2,2-dimethylbutane	19	2-3
methyl tert-butyl ether[b] +		
diethylether	22	2-4
dipentylether	21	2-5

[a] Reference [11].
[b] Reference [12].

TABLE III Excess enthalpy data of water + alkanolamine systems at 298.15 K

Systems	*Data points*	*System no.*
water[a] +		
monoethylethanolamine	16	3-1
diethylethanoleamine	18	3-2
n-propylethanolamine	17	3-3
water[b] +		
diethanoleamine	13	3-4
triethanolamine	17	3-5
methydiethanolamine	16	3-6

[a] Reference [8].
[b] Reference [9].

2.1. Chlorohydrocarbon + Hydrocarbon 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.

TABLE IV Excess enthalpy data of mono and polybromoalkanes systems at 298.15 K

Systems	*Data points*	*System no.*
tribromomethane[a] +		
1,2-dibromoethane	23	4–1
dibromoethane	17	4–2
1-bromopropane	25	4–3
1,1,2,2-tetrabromoethane[a] +		
tribromoethane	20	4–4
1,2-dibromoethane	22	4–5
dibromomethane	20	4–6
1-bromopropane	20	4–7
1,2-dibromoethane[a] +		
dibromoethane	14	4–8
1-bromopropane	22	4–9
dibromomethane[a] +		
1-bromopropane	19	4–10

[a] Reference [13].

2.2. Ether + Hydrocarbon 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.

2.3. Water + Alkanolamine Systems

The third part of the data base contains 97 data points of 6 binary mixture. The binary systems of part three are listed in Table III.

2.4. Mono and Poly Bromoalkanes Systems

The fourth part of the database contains 202 data points of 10 binary mixtures. The binary systems of part four are listed in Table IV.

3. ANALYSIS OF THE DATABASE

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

3.1. The excess enthalpy-composition plot changes sign and goes from positive to negatives values [4].

3.2. The excess enthalpy shows large negative or positive values greater than $500\,\mathrm{J\,mol^{-1}}$ [4–9].
3.3. The excess enthalpy shows moderate negative or positive values less than $500\,\mathrm{J\,mol^{-1}}$ [10–13].
3.4. The excess enthalpy-composition plot is symmetric of asymmetric [4–13].

4. DATA REDUCTION BY REDLICH-KISTER EQUATION

The Redlich-Kister equation (R-K) [14] is a flexible function commonly used for correlation binary excess enthalpy data. It is an empirical power expansion

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

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 datasets of the excess enthalpy were correlated by Redlich-Kister equation, and the results are given in the literature [4–13].

5. CORRELATION BY WILSON AND NRTL MODELS

5.1. Wilson Equation (15)

In the development of the equation, Wilson conceived that 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 a mixture 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:

$$\frac{p_{21}}{p_{11}} = \frac{x_2\exp(-a_{21}/RT)}{x_1\exp(-a_{11}/RT)} \qquad (2)$$

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

With 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 (16)

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} indicating the different interaction energies. The nonrandomness parameter α_{21} is an empirical constant.

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

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

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} was taken to be constant for each binary mixtures studied here. The adjustable parameters λ_{21} and λ_{12} in Wilson equation and Δg_{21}, Δg_{12} and α_{21} in NRTL equation were obtained by Newton-Raphson least square fit and are summarized in Tables V–VIII.

TABLE V 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 chlorohydrocarbon + hydrocarbon systems at 298.15 K

	NRTL					*Wilson*			
System no.	Δg_{12}	Δg_{12}	α_{12}	*AD*	*RD*	λ_{12}	λ_{21}	*AD*	*RD%*
1-1	−32033.2	40826.7	0.01	13.8	2.63	−2342.1	1000.1	39.4	7.48
1-2	−32960.2	42548.6	0.01	21.7	3.77	−2300.9	1257.9	33.3	5.79
1-3	−33954.3	44207.3	0.01	27.93	4.72	−2291.1	1688.1	32.1	5.43
1-4	−34403.7	44933.5	0.01	41.18	6.73	−2150.7	1397.4	28.8	4.71
1-5	−34868.6	45638.9	0.01	41.56	6.5	−2059.2	1347.2	20.8	3.25
1-6	−34586.6	44877.8	0.01	28.13	4.22	−1934.3	1123.9	18.5	2.78
1-7	−37662.9	52547.5	0.01	88.2	16.2	6341.1	−1410.6	161.8	29.7
1-8	−37419.4	51969.4	0.01	71.4	12.8	6381.7	−1658.6	136.8	24.5
1-9	−33242.4	43861.6	0.01	95.7	16.8	6670.0	−1852.4	173.4	30.5
1-10	−33029.4	43528.2	0.01	91.8	15.9	7064.4	−2130.8	170.5	29.5
1-11	−387140.0	55792.8	0.01	87.3	15.2	6922.1	−2076.6	161.8	28.1
1-12	−33141.4	43702.2	0.01	90.3	15.6	7157.7	−2401.4	151.1	26.1
1-13	156482.5	−142543.8	0.001	15.2	1.47	−327.2	−2786.1	11.8	1.14
1-14	−5716.1	10077.0	0.20	18.7	1.33	−3020.5	−968.0	63.7	4.51
1-15	−164374.9	179609.6	0.001	14.3	0.676	−3558.4	−1926.0	54.7	2.59
1-16	−31898.7	43987.2	0.022	6.75	0.199	−4588.3	−2911.7	237.7	7.01
1-17	2805	7048.2	0.25	30.3	2.71	3810.7	3097.0	292.8	26.2
1-18	932.2	6278.0	0.20	61.0	5.83	3964.5	2992.1	240.2	22.9
1-19	923.2	10061.8	0.25	28.0	3.50	5609.8	1054.7	132.1	16.5
1-20	1558.4	4913.3	0.25	56.7	5.97	3806.2	3109.3	196.5	20.7
1-21	715.7	759.7	1.70	1.83	0.968	625.0	212.6	5.08	2.69
1-22	1039.7	1005.4	1.10	1.26	0.437	967.6	320.2	4.21	1.45
1-23	1336.7	1142.4	0.80	2.91	0.768	1184.7	548.6	5.31	1.40
1-24	2448.1	1869.4	0.58	2.66	0.482	1689.9	1251.6	7.44	1.35
1-25	−78.2	461.3	0.80	2.06	2.57	545.3	−158.2	2.02	2.53
1-26	318.2	397.6	0.85	0.850	0.539	551.7	113.3	0.872	0.553
1-27	703.3	521.2	1.00	1.71	0.743	600.3	400.7	1.87	0.812

TABLE VI Adjustable parameters, Δg_{ij}, λ_{ij}, a_{12}, absolute deviation, *AD* and relative deviation, *RD* based on NRTL and Wilson models for calculating excess molar enthalpies for ether + hydrocarbon systems at 298.15 K

	NRTL					*Wilson*			
System no.	Δg_{12}	Δg_{12}	α_{12}	*AD*	*RD*	λ_{12}	λ_{21}	*AD*	*RD%*
2–1	−130.2	1221.5	0.20	1.45	0.599	827.2	297.8	1.58	0.651
2–2	−161.9	1293.7	0.20	1.70	0.678	851.5	315.4	1.65	0.659
2–3	−136.0	1135.5	0.20	1.38	0.615	766.9	258.2	1.47	0.656
2–4	−181.5	284.1	0.50	0.373	1.80	136.0	−40.2	0.377	1.82
2–5	831.4	−113.7	0.50	0.681	0.469	390.8	162.3	0.548	0.378

TABLE VII Adjustable parameters, Δg_{ij}, λ_{ij}, a_{12}, absolute deviation, *AD* and relative deviation, *RD* based on NRTL and Wilson models for calculating excess molar enthalpies for water + alkanolamine systems at 298.15 K

	NRTL					*Wilson*			
System no.	Δg_{12}	Δg_{12}	α_{12}	*AD*	*RD*	λ_{12}	λ_{21}	*AD*	*RD%*
3–1	−4571.5	−16627.8	−0.11	58.4	2.4	−4639.4	−2060.0	93.9	3.87
3–2	−4612.1	−18572.0	−0.10	76.9	2.90	−5290.0	−2120.3	107.1	4.04
3–3	−8597.8	6761.0	0.12	64.1	3.25	−4766.2	−1406.2	36.5	1.86
3–4	24136.6	−10099.0	0.09	175.9	8.43	−4124.5	−1789.8	128.1	6.14
3–5	20871.8	−9544.8	0.09	141.1	9.71	−3539.0	−1340.3	45.3	3.12
3–6	25995.7	−11025.7	0.09	342.8	13.8	−4453.6	−2174.9	144.2	5.78

TABLE VIII Adjustable parameters, Δg_{ij}, λ_{ij}, a_{12}, absolute deviation, *AD* and relative deviation, *RD* based on NRTL and Wilson models for calculating excess molar enthalpies for mono and polybromoalkanes systems at 298.15 K

	NRTL					*Wilson*			
System no.	Δg_{12}	Δg_{12}	α_{12}	*AD*	*RD*	λ_{12}	λ_{21}	*AD*	*RD%*
4–1	2417.7	−2345.2	0.20	1.08	0.529	−656.3	−76.2	1.10	0.537
4–2	622.1	−391.4	2.0	0.671	1.40	−1261.5	1569.4	0.876	1.83
4–3	2245.8	−1540.4	0.50	3.75	1.93	−1577.3	2865.7	4.63	2.39
4–4	−876.9	1118.3	0.20	0.290	1.38	430.8	−215.0	0.360	1.71
4–5	−2768.9	3290.6	0.20	2.65	1.11	−791.8	−125.8	2.68	1.12
4–6	−306.7	472.3	4.0	1.12	1.42	−1835.4	2061.8	1.49	1.88
4–7	2099.9	−1272.5	0.60	2.16	3.07	−1788.0	2724.0	2.80	1.97
4–8	774.5	−664.3	0.50	0.300	1.20	−104.9	−2.60	0.321	1.29
4–9	803.4	625.9	0.50	3.59	1.17	673.1	53.8	3.57	1.16
4–10	1616.6	−784.6	0.20	0.726	0.497	89.4	629.6	0.984	0.674

TABLE IX Parameters of modified NRTL equation for binary mixtures of trichloroethylene in akan-1-ols at 298.15 K

Parameters	1-*propanol*	1-*butanol*	1-*pentanol*	1-*hexanol*	1-*heptanol*	1-*octanol*
Δg_{12}	-37662.9	-37419.4	-33242.3	-33029.4	-38714.0	-33141.1
Δg_{12}	52547.5	51969.4	43861.6	43528.2	55792.8	43702.2
B_1	9.78666×10^{-3}	9.83266×10^{-3}	9.73419×10^{-3}	9.78517×10^{-3}	9.91923×10^{-3}	9.87602×10^{-3}
B_2	1.55281×10^{-4}	1.77773×10^{-4}	-9.32454×10^{-4}	-6.14002×10^{-4}	1.12482×10^{-4}	-7.07277×10^{-4}
B_3	-4.79365×10^{-4}	-4.72267×10^{-4}	4.37571×10^{-3}	5.99132×10^{-3}	2.39455×10^{-3}	1.67598×10^{-4}
B_4	2.39401×10^{-3}	1.64610×10^{-3}	3.44806×10^{-3}	2.60544×10^{-3}	2.64660×10^{-3}	1.98313×10^{-3}
B_5	3.41279×10^{-3}	3.29088×10^{-3}	-1.29220×10^{-2}	-1.87622×10^{-2}	-5.89386×10^{-3}	3.99394×10^{-3}
B_6	-4.97259×10^{-3}	-4.05409×10^{-3}	-7.26600×10^{-3}	-6.64918×10^{-3}	-4.37604×10^{-3}	-5.31947×10^{-3}
B_7			1.49808×10^{-2}	2.00018×10^{-2}	7.49742×10^{-3}	
RD%	3.59	2.7	3.48	4.4	2.55	2.48
AD	19.6	15.1	19.8	25.4	14.7	14.3

6. MODIFIED NRTL EQUATION

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

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

where B_i is 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) and the same Δg_{21}, Δg_{12} were calculated for NRTL equation, we used Tables V–VIII, and experimental excess enthalpy for each mole fraction and found α_{12} for minimum deviation between experimental and calculated excess enthalpy. Finally the parameters of Eq. (6) were obtained by least square fit.

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

7. RESULTS AND DISCUSSION

The results of data correlation by Wilson and NRTL models for 48 selected systems are given in Tables V–VIII for each systems absolute deviation, *AD* and relative deviation, *RD* based on following equation

TABLE X Parameters of modified NRTL equation for binary mixtures of water in alkanolamines at 298.15 K

Parameters	*Diethanolamine*	*Triethanolamine*	*Methyldiethanolamine*
Δg_{12}	24136.8	20871.8	25995.7
Δg_{21}	-10099.0	-9544.8	-11025.7
B_1	8.62596×10^{-2}	8.23426×10^{-2}	8.51828×10^{-2}
B_2	1.15954×10^{-2}	1.11504×10^{-2}	-7.29432×10^{-3}
B_3	3.57780×10^{-2}	5.06016×10^{-2}	-674062×10^{-2}
B_4	-3.45187×10^{-2}	-4.30475×10^{-4}	1.40595×10^{-1}
B_5	-3.33793×10^{-2}	-9.96875×10^{-2}	4.63199×10^{-1}
B_6	6.63303×10^{-2}	4.02183×10^{-2}	-8.65126×10^{-2}
B_7	9.13218×10^{-2}	1.56965×10^{-1}	-3.57984×10^{-1}
RD%	0.344	0.896	4.8
AD	7.17	13.0	119.7

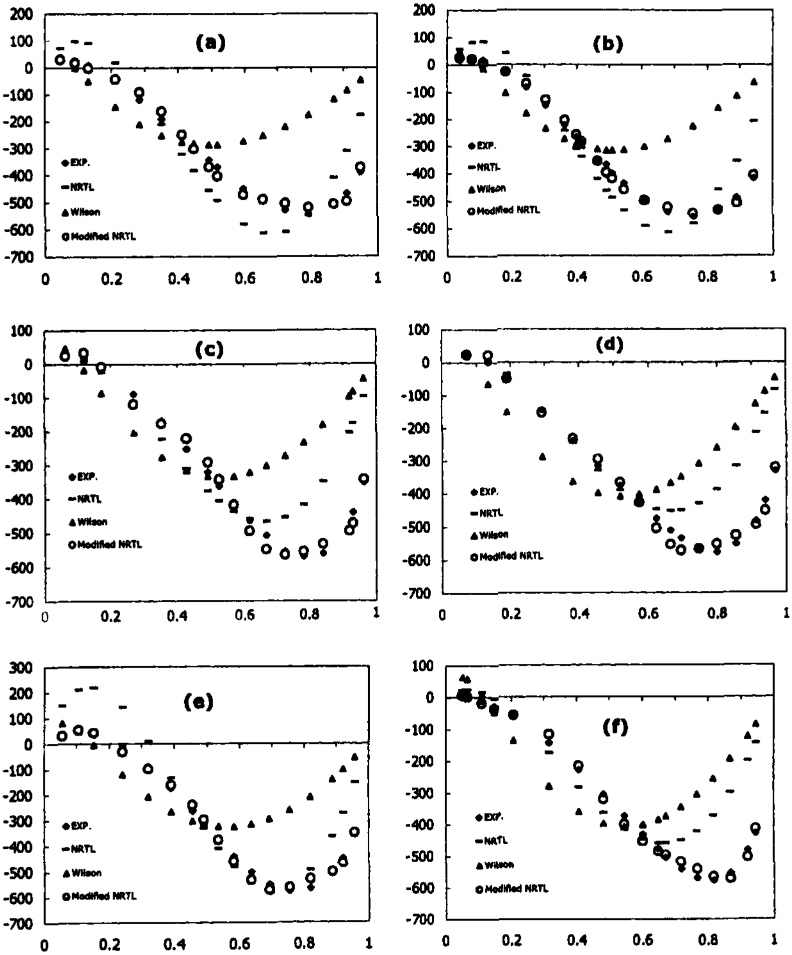

FIGURE 1 Comparison between calculated [Wilson Equation (3), NRTL Equation (5) and modified NRTL Equations (5) and (6)] using coefficients from Tables V and IX and experimental $H_m^E/\mathrm{J\,mol^{-1}}$ values (Y) for (a) 1-propanol, (b) 1-butanol, (c) 1-pentanol, (d) 1-hexanol, (e) 1-heptanol and (f) 1-octanol + (X) mole fraction of trichloroethylene at 25°C.

were calculated

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

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

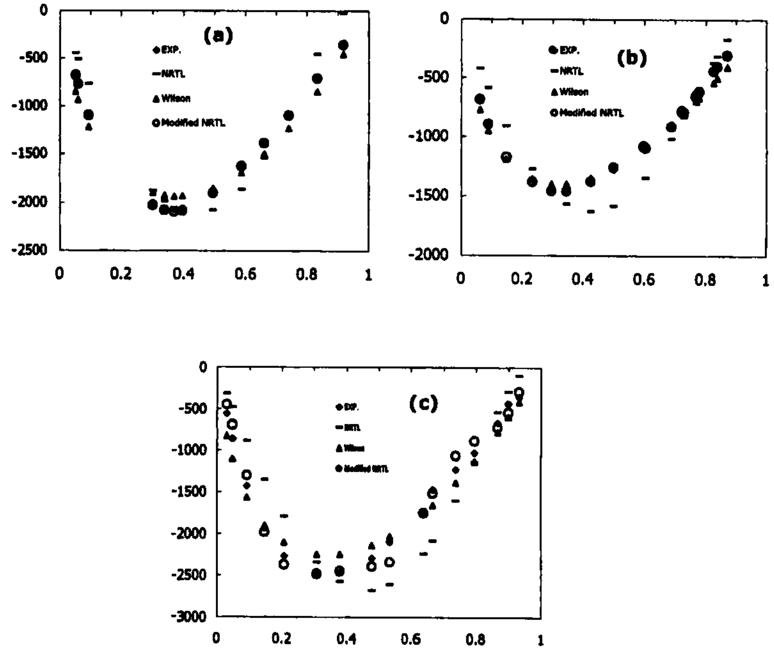

FIGURE 2 Comparison between calculated [Wilson Equation (3), NRTL Equation (5) and modified NRTL Equations (5) and (6)] using coefficients from Tables VII and X and experimental $H_m^E/\mathrm{J\,mol^{-1}}$ values (*Y*) for (a) diethanolamine, (b) triethanolamine and (c) methyldiethanolamine + water. (*X*) is mole fraction of first component, respectively, at 25°C.

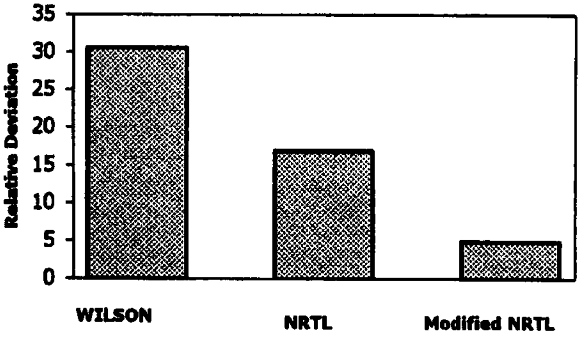

FIGURE 3 Overall deviation by different models.

where N is the number of data point and $H^E_{(\exp)_{\max}}$ is the maximum experimental excess enthalpy absolute in each systems. For nine systems that deviations are relatively large, trichloroethylene + alkan-1-ols [4] and water + alkanolamines [9] the modified NRTL model were applied and the results are summarized in Tables IX and X. Figures 1 and 2 show comparison of the experimental and theoretical H^E_m values as a function of mole fraction derived from Wilson, NRTL and modified NRTL equations and Figure 3 shows the variation of relative deviation by different models for all systems. The maximum number of adjustable parameters used in the Redlich-Kister equation is 9 and the best results were obtained. On the other hand, fewer parameters are necessary for correlation with Wilson, NRTL and modified NRTL models for modified NRTL model the relative deviation is smaller than Wilson and NRTL and comparable with Redlich-Kister equations.

In cases when excess enthalpies are not large and $H^E_m - x$ curves are symmetric, all tested models are able to fit the experimental data with good accuracy. Among them NRTL equation provides the best fits for such systems. However, when excess enthalpy curve is asymmetric the modified NRTL model is better.

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