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MODELING AND EXPERIMENTAL DATA VALIDATION OF VAPOR LIQUID EQUILIBRIA (VLE) FOR ABSORPTION AND DISTILLATION

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ABSTRACT. The model combing the method UNIFAC with various model methods was applied for calculation of activity coefficients in an VLE model for hexaflourobenzene(1) benzene(2) at 333.15K, butanol(1) tetrachloroethene(2) at 101.08 kPa, carbondioxide(1) 3-pentanol(2) at 313.2K and other system compounds. The simulated results represent the overall average relative deviation below 0.5% which is comparable to the results presented by other authors in the literature. The data and results obtained for the systems are compared with the compositions predicted by the model and the sensitivity of the model parameters towards the composition data was analyzed through simulation techniques. The results predicted that overall error convergence was found to be less than 0.5 %.

Keywords: *Activity coefficient, VLE, Models, convergence*.

INTRODUCTION

A number of thermodynamic models have been proposed in different literatures. VLE models are based on fundamental equations for phase and chemical equilibria [1] and differ in the activity coefficient model is reported [2]. Since the EOS [3] for the calculation of fugacity coefficient in the liquid phase is relatively well established, the activity coefficients were identified as the key variables of the VLE models [4, 5]. Rigorous modeling and simulation of activity coefficients in this type of compounds is a challenge because the physical phenomena to be described as complex. In addition, the parameter regression is found to be difficult. The recent modeling is successful in data representation and interpretation of experimental data. However, the optimal approach is still uncertain except for reliable predictions. However, most of the mixtures encountered industrially are non – ideal. The non-ideality can be described by any of the several G^E models and EOS methods [6-8].

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THERMODYNAMIC PROPERTY METHODS FOR MODELING

ACTIVITY COEFFICIENT MODELS

In the literature several activity coefficient models are proposed and the oldest of the equations still in common use is that of Margules [9].

$$\left(\frac{G^E}{RT}\right)_{CAL} = x_1 x_2 \left[A_{21} x_1 + A_{12} x_2\right] \tag{1}$$

Van Laar equations are originally based on the Van der Waals equation of state but now these are regarded as pure empirical equations. Margules and Van Laar models can be expressed in linear form.

$$\left(\frac{G^E}{RT}\right)_{CAL} = \left[\frac{A'_{12} A'_{21}}{A'_{12} X_1 + A'_{21} X_2}\right] X_1 X_2 \tag{2}$$

Linear form of Van Laar model [10]

$$\left(\frac{G^E}{RT}\right)_{CAL} = \frac{1}{\left(\frac{1}{x_1 A_{12}} - \frac{1}{x_2 A_{21}}\right)} \tag{3}$$

Three parameter form of Van Laar equation [10]

$$\left(\frac{G^E}{RT}\right)_{CAL} = \frac{1}{\left(\frac{1}{x_1 A_1} - \frac{1}{x_2 A_2}\right)} + A_3 x_1 x_2 \left(x_1^2 - x_2^2\right) \tag{4}$$

Power series expression for the excess Gibbs energy and the corresponding activity coefficients for three parameter model [9] represented below.

$$\left(\frac{G^E}{RT}\right)_{CAL} = X_1 X_2 \left[B + C\left(X_1 - X_2\right)\right] \tag{5}$$

$$\left(\frac{G^E}{RT}\right)_{CAL} = x_1 x_2 \left[A_1 + A_2\left(x_1 - x_2\right) + A_3\left(x_1 - x_2\right)^2\right] \tag{6}$$

The model developed using volume fractions as measures of composition [11].These equations appear to take into account an important difference between molecules and should possibly have received more attention.

$$\Phi_1 = \frac{X_1 V_1}{X_1 V_1 + X_2 V_2} \quad (7) \qquad \Phi_2 = \frac{X_2 V_2}{X_1 V_1 + X_2 V_2} \quad (8)$$

$$\left(\frac{G^E}{RT}\right)_{CAL} = \Phi_1 \Phi_2 (X_1 V_1 + X_2 V_2)[A_{12} + (\Phi_1 - \Phi_2) A_{21}] \quad (9)$$

Equation for excess Gibbs–energy [11].

$$\left(\frac{G^E}{RT}\right)_{CAL} = x_1 x_2 A_{12} \frac{(\partial_1 - \partial_2)^2}{RT\left(\frac{1}{V_1 x_1} + \frac{1}{+V_2 x_2}\right)} \quad (10)$$

Expression for the excess Gibbs free energy [12] and substituting local compositions in the Flory– Huggins equations for polymer solutions. Wilson equations are not suitable for partially miscible systems [13]

$$\Lambda_{12} = \frac{V_2}{V_1} EXP\left(-\frac{A_{12}}{RT}\right) \quad (11) \qquad \Lambda_{21} = \frac{V_1}{V_2} EXP\left(-\frac{A_{21}}{RT}\right) \quad (12)$$

$$\left(\frac{G^E}{RT}\right)_{CAL} = -X_1 \ln(X_1 + \Lambda_{12} X_2) - X_2 \ln(X_2 + \Lambda_{21} X_1) \quad (13)$$

$$\tau_{21} = \frac{A_{21}}{RT} \quad (14) \qquad \tau_{12} = \frac{A_{12}}{RT} \quad (15) \qquad G_{12} = EXP(\tau_{12}) \quad (16) \qquad G_{21} = EXP(\tau_{21}) \quad (17)$$

Flory and Huggins equation

$$\Phi_1 = \frac{x_1 V_1}{x_1 V_1 + x_2 V_2} \quad (18) \qquad \Phi_2 = \frac{x_2 V_2}{x_1 V_1 + x_2 V_2} \quad (19)$$

$$\left(\frac{G^E}{RT}\right)_{CAL} = x_1 x_2 \left[\frac{G_{21} \ln(G_{21})}{x_1 + x_2 G_{21}} + \frac{G_{12} \ln(G_{12})}{x_2 + x_1 G_{12}}\right] \quad (20)$$

Studied the properties of Wilson equation and the equation has been the starting point for a class of activity coefficient equations collectively known as local–composition equations [14].

$$\Lambda_{12} = \frac{V_2}{V_1} EXP\left(-\frac{A_{12}}{RT}\right) \quad (21) \qquad \Lambda_{21} = \frac{V_1}{V_2} EXP\left(-\frac{A_{21}}{RT}\right) \quad (22)$$

$$\left(\frac{G^E}{RT}\right)_{CAL} = X_1 \ln\left(\frac{1}{X_1 + \Lambda_{12}X_2}\right) + X_2 \ln\left(\frac{1}{X_2 + \Lambda_{21}X_1}\right) - \frac{X_1X_2\Lambda_{12}\ln(\Lambda_{12}\Lambda_{21})}{X_1 + \Lambda_{12}X_2} \quad (23)$$

The Non Random Two Liquid (NRTL) equation using Scott's two liquid theories and taking into account non–randomness of mixing are follows [15, 16].

$$\Lambda_{12} = \frac{V_2}{V_1}\mathrm{EXP}\left(-\frac{A_{12}}{RT}\right) \quad (24) \qquad \Lambda_{21} = \frac{V_1}{V_2}\mathrm{EXP}\left(-\frac{A_{21}}{RT}\right) \quad (25)$$

$$\left(\frac{G^E}{RT}\right)_{CAL} = X_1 \ln(X_1 + X_2\Lambda_{21}) + X_2 \ln(X_2 + X_1\Lambda_{12}) \quad (26)$$

Modified form of Wilson equation by combining Flory–Huggins term and Wilson equation and incorporating molar volume ratios

$$\Lambda_{12} = \frac{V_2}{V_1}\mathrm{EXP}\left(-\frac{A_{12}}{RT}\right) \quad (27) \qquad \Lambda_{21} = \frac{V_1}{V_2}\mathrm{EXP}\left(-\frac{A_{21}}{RT}\right) \quad (28)$$

$$\Phi_1 = \frac{X_1R_1}{X_1R_1 + X_2R_2} \quad (29) \qquad \Phi_2 = \frac{X_2R_2}{X_1R_1 + X_2R_2} \quad (30)$$

$$\left(\frac{G^E}{RT}\right)_{CAL} = X_1 \ln\left(\frac{\Phi_1}{X_1}\right) + X_2 \ln\left(\frac{\Phi_2}{X_2}\right) + X_1 \ln\left(\frac{X_1R_1 + X_2R_2}{X_1R_1 + X_2R_2\Lambda_{12}}\right) + X_2 \ln\left(\frac{X_1R_1 + X_2R_2}{X_2R_2 + X_1R_1\Lambda_{21}}\right) \quad (31)$$

The UNIQUAC (Universal Quasi–chemical) equation based on Guggenheim's quasi–chemical analysis generalized through introduction of the local area fraction as the primary concentration variable [17, 18].

$$\tau_{12} = \mathrm{EXP}\left(-\frac{A_{12}}{RT}\right) \quad (32) \qquad \tau_{21} = \mathrm{EXP}\left(-\frac{A_{21}}{RT}\right) \quad (33) \qquad \Phi_1 = \frac{X_1R_1}{X_1R_1 + X_2R_2} \quad (34)$$

$$\Phi_2 = \frac{X_2R_2}{X_1R_1 + X_2R_2} \quad (35) \qquad \theta_1 = \frac{X_1Q_1}{X_1Q_1 + X_2Q_2} \quad (36) \qquad \theta_2 = \frac{X_2Q_2}{X_1Q_1 + X_2Q_2} \quad (37)$$

$$\left(\frac{G^E}{RT}\right)_{CAL} = X_1 \ln\left(\frac{\Phi_1}{X_1}\right) + X_2 \ln\left(\frac{\Phi_2}{X_2}\right) - \left(\frac{Z}{2}\right)\left[X_1 Q_1 \ln\left(\frac{\theta_1}{\Phi_1}\right) - X_2Q_2 \ln\left(\frac{\theta_2}{\Phi_2}\right)\right] - X_1 Q_1 \ln(\theta_1 + \theta_2\tau_{21}) - X_2 Q_2 \ln(\theta_2 + \theta_1\tau_{12}) \quad (38)$$

Suggested modification in the Wilson equation to give weightage to the molar volumes of the components are given below [19].

$$\tau_{12} = \frac{V_2}{V_1} EXP\left(-\frac{A_{12}}{RT}\right) \quad (39) \qquad \tau_{21} = \frac{V_1}{V_2} EXP\left(-\frac{A_{21}}{RT}\right) \quad (40)$$

$$\left(\frac{G^E}{RT}\right)_{CAL} = X_1 \ln\left(\frac{X_1 + \left(\frac{V_2}{V_1}\right)X_2}{X_1 + X_2\tau_{12}}\right) + X_2 \ln\left(\frac{X_2 + \left(\frac{V_1}{V_2}\right)X_1}{X_2 + X_1\tau_{21}}\right) \quad (41)$$

The following is the two forms to represent the VLE data of alcohol – hydrocarbon mixtures obtained at 313.15 K [20 - 23]

$$\left(\frac{G^E}{RT}\right)_{CAL} = \frac{x_1 x_2}{A_{12} + A_{21}(2x_1 - 1)} \quad (42)$$

PREDICTIVE MODELS

Estimation of thermodynamic properties of liquid mixtures from group contributions was first suggested [24] used group contributions to correlate heats of mixing. (a) UNIFAC (UNIQUAC Functional group Activity Coefficients) (b) Derr and Deal (1969) extended the Analytical Solution of Groups (ASOG) method (c) Equations of state.

(i) **Equation of State for Simple Molecules**

The van der Waals equation of state was the first equation capable of representing vapour-liquid coexistence [25, 26]

$$P = \frac{RT}{V - b} - \frac{a}{V^2} \quad (43)$$

The van der Waals equation can be regarded as a “hard-sphere (repulsive) + attractive” term equation of state composed from the contribution of repulsive and attractive intermolecular interactions, respectively.

(ii) **PARAMETER ESTIMATION**

The estimation parameters in the G^E models [27] and EOS methods had received wide attention since the parameters in a given model contain concisely the information on the composition dependent behavior [28] of the solution. The following methods appear to be more popular. The popular

optimization methods are Genetic Algorithm, Simulated Annealing Goldstein and Price, Greenstadt and Goldfarb, Approximation of Hessian, Zoutendijk's. Hook and Jeeves, Neldermead Simplex Method

(iii) **MODEL VALIDATION**

Activity Coefficient Models [29, 30] **Algorithm**

Step 1. Read P_c, V_c, T_c, ω, R, Q (UNIQUAC) for components 1 and 2
Step 2. Read Antoine constants A, B, C for components 1 and 2. Read T, P, x, y data and number of point's 'n'
Step 3. Calculate saturated molar volume V for components 1 and 2 using Racket equation
Step 4. Calculate using Tsonpolous correlation for components 1 and 2 by assuming vapor phase is ideal
Step 5. Minimize the objective function using optimization methods
Step 6. Calculate the absolute average deviation

EOS Models Algorithm for Peng Robinson and Margules mixing Rule

Step1. Read T_c, P_c, ω for components 1 and 2
Sgtep 2. Read T, P, x, y data. Find the values of the mixing rules using Margules Equation
Step 3. The below function is minimized using Hooke & Jeeves optimization method
Step 4. The optimum values of k_1, k_2 and k_3 are found
Step 5. The predicted vapor composition can be evaluated using the following correlation [31 - 34]

$$\ln\phi_i^V = \frac{b_i}{b_{vap}}\left(Z_{vap}-1\right)-\ln\left(Z_{vap}-\beta_{vap}\right)-\frac{q_{vap}}{2\sqrt{2}}\left(\frac{2\sum_{j=1}^{c}x_j a_{ij}}{a_{vap}}-\frac{b_i}{b_{vap}}\right)I_V \qquad (44)$$

RESULTS AND DISCUSSION

The modeling and simulation of VLE models (Table 1, 2, 3, and 4) for various chemical compounds are studied and are compared with the experimental results. The results predict that error of convergence is less than 0.5%. The phase diagrams [35] are drawn by plotting the data of experimental and model predicted data, which are shown in the figure 1.

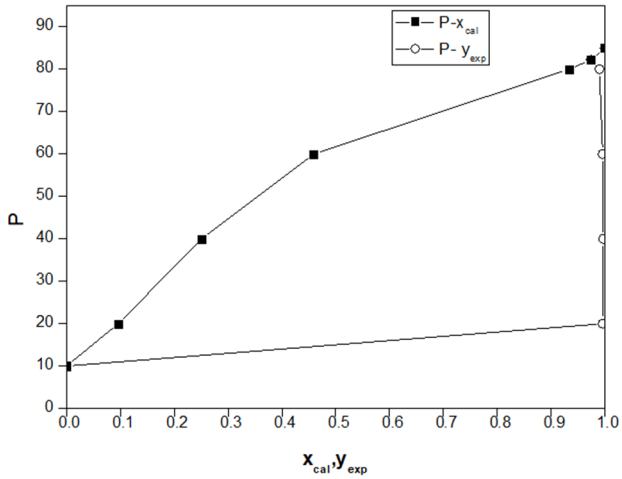

Figure 1. P-X-Y plot for experimental and predicted given in Table 4

Table 1. Objective functions for activity coefficient models and equation of state models

Objective functions for activity coefficient models	
$\sum_{i}^{n}\left[\frac{G^{E}_{EXP}}{RT}-\frac{G^{E}_{CAL}}{RT}\right]^{2}$ (45)	$\sum_{i}^{n}\left[\left(\frac{(\gamma_{1EXP}-\gamma_{1\,CAL})}{(\gamma_{1EXP})}\right)^{2}+\left(\frac{(\gamma_{2EXP}-\gamma_{2CAL})}{(\gamma_{2EXP})}\right)^{2}\right]$ (46)
$\sum_{i}^{n}\left[(\gamma_{1EXP}-\gamma_{1\,CAL})^{2}+(\gamma_{2EXP}-\gamma_{2CAL})^{2}\right]$ (47)	$\sum_{i}^{n}\left[(y_{EXP}-y_{CAL})^{2}+(P_{EXP}-P_{CAL})^{2}\right]$ (48)
$\sum_{i}^{n}(y_{EXP}-y_{CAL})^{2}$ (49)	$\sum_{i}^{n}(P_{EXP}-P_{CAL})^{2}$ (50)
Objective functions for equation of state	
$\sum_{i}^{n}\left[(\varphi_{1}^{vap}*y_{1}*P-\varphi_{1}^{liq}*x_{1}*P)^{2}+(\varphi_{2}^{vap}*y_{2}*P-\varphi_{2}^{liq}*x_{2}*P)^{2}\right]$ (51)	

Table 2. Component system 1: Hexafluorobenzene (1) Benzene (2) at 333.15 K

P_{exp}, atm	x	y_{exp}	ycal				
			Ideal vapor	Correlation	Prediction	PR-EOS	SRK-EOS
			NELD-MCLC	ZOUT-NRTL	POLA-UNQ1	SWTR-PRSV-7	MARG-HAKP-7
0.52160	0.0000	0.0000	0.000000	0.000000	0.000000	0.000000	0.000000
0.52570	0.0941	0.0970	0.096541	0.095746	0.096470	0.097118	0.097109
0.52568	0.1849	0.1788	0.177904	0.177742	0.177621	0.178485	0.178546
0.52287	0.2741	0.2567	0.255947	0.256227	0.255765	0.256388	0.256417
0.51818	0.3648	0.3383	0.338211	0.338493	0.338251	0.338498	0.338394
0.50989	0.4538	0.4237	0.424064	0.423824	0.423828	0.424228	0.424015
0.50773	0.5266	0.4982	0.498493	0.498231	0.498175	0.498610	0.498415
0.50350	0.6013	0.5783	0.578339	0.577842	0.577462	0.578438	0.578412
0.49974	0.6894	0.6760	0.675496	0.674796	0.673922	0.675604	0.675888
0.49757	0.7852	0.7824	0.781704	0.780786	0.779786	0.781831	0.782265
0.49794	0.89600	0.8996	0.899691	0.898726	0.898377	0.899793	0.899623
0.50155	1.0000	1.0000	1.000000	1.000000	1.000000	1.000000	1.000000
ΔY			0.000349	0.000624	0.000818	0.000265	0.000138

Table 3. Component system 2–Butanol (1) Tetracholoroethene (2) at 101.08 kPa

T_{exp}, K	x	y_{exp}	y_{cal}			
			Correlation	Prediction	PR-EOS	SRK-EOS
			POWE-SCHA	PNO2-MALA	SWTR-BOUM-7	SWTR-PARE-9
394.25	0.0000	0.0000	0.000000	0.000000	0.000000	0.000000
387.65	0.0380	0.2050	0.192768	0.193936	0.204900	0.204809
380.25	0.1170	0.4030	0.400829	0.401654	0.403970	0.404597
376.65	0.1820	0.4910	0.491572	0.491092	0.489515	0.489859
374.05	0.2630	0.5590	0.561726	0.560027	0.559623	0.558700
372.55	0.3480	0.6100	0.609357	0.607066	0.610110	0.608314
371.35	0.4720	0.6620	0.655817	0.653841	0.659451	0.658476
370.95	0.5200	0.6720	0.670137	0.668674	0.673110	0.673327
370.55	0.5680	0.6820	0.683975	0.682994	0.685076	0.686802
370.35	0.6120	0.6970	0.697293	0.696545	0.697287	0.699782
370.25	0.6620	0.7130	0.713689	0.713194	0.712832	0.715683
370.25	0.8240	0.7880	0.791115	0.790386	0.785333	0.787435
370.75	0.8850	0.8360	0.840070	0.838996	0.835602	0.835619
371.35	0.9330	0.8890	0.892802	0.891630	0.890318	0.888928
371.65	0.9700	0.9460	0.945899	0.945015	0.944175	0.942971
372.45	0.9920	0.9840	0.984445	0.984151	0.984374	0.983774
372.65	1.0000	1.0000	1.000000	1.000000	1.000000	1.000000
ΔY			0.002459	0.002333	0.001004	0.001430

Table 4. Component system 3: Carbon dioxide (1) 3–Pentanol (2) at 313.2 K

P_{exp}, bar	x_{cal}	y_{exp}	y_{cal}	
			PR-EOS	SRK-EOS
			MARG-PRSV -4	MARG-THRO-5
10.0000	0.00000	0.0000	0.000000	0.000000
20.0000	0.09600	0.9960	0.996000	0.996000
40.0000	0.25100	0.9970	0.997000	0.997000
60.0000	0.45900	0.9960	0.996000	0.996000
80.0000	0.93300	0.9900	0.989999	0.989999
82.2000	0.97400	0.9740	0.974000	0.974000
85.0000	1.00000	1.0000	1.000000	1.000000
AAD in y			0.000000	0.000000

Table 5. Different component systems property model data

System Name	Temp/Pres	Δy		
		PR–EOS	DECHEMA	PR – Mathias
Methanol(1) Acetonitrile(2)	303.15 K	0.005162	0.0043(Uniq)	0.005097
Methanol(1) 1,2–Dichloroethane(2)	323.15 K	0.004235	0.0031(Marg)	0.000776
2–Methyl1propanol(1) 3–Methylbutanol(2)	353.15 K	0.007487	0.0090(NRTL)	0.002130
Propionicaldehyde(1) Methanol(2)	318.15 K	0.005853	0.0058(Uniq)	0.003616
Benzene(1) 1–Butanol(2)	1.0132 bar	0.003308	0.0023(Vanl)	0.001972
Ethylacetate(1) 2–Methoxyethanol(2)	1.0132 bar	0.004412	0.0043(Wils)	0.004190
Ethanol(1) Ethylbenzene(2)	1.0132 bar	0.011148	0.0026(NRTL)	0.000535
Ethanol(1) Tetrachloroethylene(2)	1.0132 bar	0.007672	0.0055(Uniq)	0.003978

NOMENCLATURE

G^E – Gibbs free energy
A – Activity coefficients
T – Temperature °C
R – Gas constant
X – Vapor moles
γ- Phase coefficients
p – Pressure, kpa
B, C, k, ω, Ø – constants
Λ – Excess free energy mixing
V – Molar volume ratios

CONCLUSIONS

In this study, model combing the method UNIFAC with various model methods was applied for calculation of activity coefficients in an VLE model for hexaflourobenzene(1) benzene(2) at 333.15K, butanol(1) tetrachloroethane (2) at 101.08 kPa, carbon dioxide(1) 3-pentanol(2) at 313.2K and other system compounds. The relative deviation between predicted and experimental values is below 0.5% error, which shows the comparable to the results given in paper with the literature available. The data and results obtained for the systems are compared with the compositions predicted by the model and the sensitivity of the model parameters towards the composition data was analyzed through simulation techniques. The results predicted that overall error convergence was found to be less than 0.5 % and commendable results obtained.

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