

# A modification of Wong-Sandler mixing rule for the prediction of vapor-liquid equilibria in binary asymmetric systems

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(Received 8 November 2010 • accepted 31 December 2010)

**Abstract**—Systems consisting of light components and heavy hydrocarbons are highly asymmetric and industrially important. Design and control of facilities for separation and purification of such mixtures require vapor-liquid equilibrium data. Coupling of the cubic equation of state (EOS) with excess Gibbs energy models (EOS/ $G^{\text{ex}}$  models) failed to represent the vapor-liquid equilibria (VLE) of such systems accurately. The main purpose of this work is to present a modification of Wong-Sandler mixing rule with using the composition dependent binary interaction parameter. Vapor-liquid equilibria for 30 binary systems are calculated using the SRK equation of state with proposed model and Wong-Sandler mixing rule. Calculated pressures and mole fractions of vapor phase are compared with experimental data. The average absolute percentage deviation indicates that error involved in the application of modified Wong-Sandler model is less than Wong-Sandler model in most cases.

Key words: Vapor-liquid Equilibrium, Wong-Sandler Mixing Rule, Asymmetric Systems

## INTRODUCTION

Differences in size, shape, free volume and molecular interactions are all responsible for non-idealities in mixtures. Size-asymmetric systems containing large and small molecules are important due to their applications in the oil and petrochemical industries. The estimation of the equilibrium phase behavior of such systems over broad ranges of temperature and pressure is an important problem in chemical process design. Many efforts have been made to represent the vapor-liquid equilibrium (VLE) of asymmetric systems accurately, but this is rather difficult because of the size difference between the components.

Equations of state are considered powerful tools for modeling of VLE. In particular, cubic equations are widely used due to their algebraic simplicity and, in many cases, their accuracy. There are two requirements for these equations to be successful: they must precisely predict the saturation pressure of pure substances and mixing rules must be available that correctly extend these equations to multi component mixtures. There has been significant improvement in the accuracy of cubic equations of state for the prediction of pure component properties [1]. The extension of equations of state to describe phase behavior for a broad range of multi component mixtures has been more difficult due to limited applicability of the van der Waals (VdW) one-fluid mixing and combining rules that are commonly used.

Wong and Sandler developed a mixing rule which combines an EOS with a free-energy model that shows the desired behavior at low and high densities without being density dependent by using the existing table of  $G^{\text{ex}}$  parameters. The Wong-Sandler model could be used to extrapolate successfully experimental data from low tem-

peratures and pressures to much higher temperatures and pressures. In the absence of experimental data, this model can be used for providing high quality predictions based on the UNIFAC model or other low pressure VLE prediction methods [2-5], but it has been shown that the Wong-Sandler model fails to represent asymmetric systems [6].

In this work, the Wong-Sandler mixing rule is modified to predict vapor-liquid equilibrium in non-ideal and asymmetric systems accurately.

## THEORY

In current study, the following form was used for the two-parameter cubic equation of state:

$$P = \frac{RT}{\underline{V} - b} - \frac{a}{(\underline{V} + ub)(\underline{V} + wb)} \quad (1)$$

To apply this equation to mixtures, the parameters  $a$  and  $b$  are considered as functions of the component compositions using mixing rules. According to the Wong-Sandler mixing rule,

$$b_m = \frac{Q}{(1-D)} \quad (2)$$

$$a_m = Q \frac{Q}{(1-D)} \quad (3)$$

$$D = \sum_i x_i \frac{a_i}{b_i RT} + \frac{A^{\text{ex}}}{CRT} \quad (4)$$

and

$$Q = \sum_i \sum_j x_i x_j \left( b - \frac{a}{RT} \right)_{ij} \quad (5)$$

with

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$$\left(b - \frac{a}{RT}\right)_{ij} = \frac{\left(b_i - \frac{a_i}{RT}\right) + \left(b_j - \frac{a_j}{RT}\right)}{2} (1 - k_{ij}) \quad (6)$$

where  $C$  is an EOS-dependent constant (VdW:  $C=-1$ , SRK:  $C=-\ln 2$ , PR:  $C=-0.623$ ),  $A_{\infty}^{\text{ex}}$  is any suitable molar excess Helmholtz free energy model at infinite pressure or equivalently an excess Gibbs free energy model at low pressures and the single adjustable parameter ( $k_{ij}$ ) for each binary pair is referred to as the WS mixing rule parameter ( $k_{12}$ ).

In the original Wong-Sandler mixing rule, the  $k_{ij}$  term could be determined by matching the Gibbs free energy predicted by the equation of state to the Gibbs free energy calculated by the original Gibbs free energy model at a specific temperature, pressure and composition, but in this work, in order to modify Wong-Sandler mixing rule, we proposed that  $k_{ij}$  must be composition dependent. To this purpose, we select the following form for  $k_{ij}$ :

$$k_{ij} = K_{ij}x_i + K_{ji}x_j \quad (7)$$

To use Eq. (7) in the WS model, we require to compute two parameters  $K_{ij}$  and  $K_{ji}$ , aforementioned in the above equation. We calculate these parameters using the relation between the activity coefficient and fugacity coefficients obtained from an EOS. The activity coefficient of species  $i$  in a mixture can be obtained from an EOS using the relation

$$\gamma_i(T, P) = \frac{\bar{\phi}_i(T, P, x_i)}{\phi_i(T, P)} \quad (8)$$

where  $\bar{\phi}_i$  is the fugacity coefficient of component  $i$  in the mixture and  $\phi_i$  is the pure component fugacity, both computed at the temperature and pressure of the mixture with an equation of state.

Consequently, the two interaction parameters per binary can be related to the activity coefficient over the whole composition range or to the values at a specified composition, such as the two infinite dilution activity coefficients of a mixture.

By writing Eq. (8) for a cubic EOS for the two infinite dilution limits, we derive the two parameters  $K_{ij}$  and  $K_{ji}$  in terms of the pure component EOS parameters and infinite dilution activity coefficient. For Eq. (1) we have

$$K_{ij} = 1 - \frac{S_j}{\left(b_i - \frac{a_i}{RT}\right) + \left(b_j - \frac{a_j}{RT}\right)} \quad (9)$$

where

$$S_j = \frac{\ln \gamma_i^{\infty} + \ln \bar{\phi}_i^{\infty}(T, P_i^{\text{vap}}) + Q_j \left(b_i - \frac{a_i}{RT}\right) + b_i \left(1 - \frac{\ln \gamma_i^{\infty}}{C} - \frac{a_j}{b_j RT}\right)}{Z_i - 1} \quad (10)$$

with

$$Q_j = \ln \frac{(V_i - b_i)Z_i}{V_i} - \frac{1}{w - u} \left( \frac{a_i}{b_i RT} + \frac{a_j}{b_j RT} + \frac{\ln \gamma_i^{\infty}}{C} \right) \ln \frac{V_i + ub_i}{V_i + wb_i} \quad (11)$$

**Table 1. Vapor-liquid equilibria of CO<sub>2</sub>+alkanes using the SRK equations of state with WS and MWS mixing rules**

System	T [K]	NP	$\Delta P/P$		$\Delta y$		Data ref.
			WS	MWS	WS	MWS	
CO <sub>2</sub> +C <sub>1</sub>	230.00-270.00	52	2.8	1.6	1.45	0.80	[12]
CO <sub>2</sub> +C <sub>2</sub>	207.00-270.00	84	0.4	0.8	2.82	1.92	[12]
CO <sub>2</sub> +C <sub>3</sub>	311.05-361.15	59	3.5	1.1	1.81	1.25	[13]
CO <sub>2</sub> +n-C <sub>4</sub>	277.90-418.48	96	1.1	0.7	2.35	1.50	[14]
CO <sub>2</sub> +n-C <sub>5</sub>	252.67-458.54	108	3.8	1.4	0.35	1.30	[15]
CO <sub>2</sub> +n-C <sub>6</sub>	298.15-313.15	20	3.4	2.9	0.65	0.34	[16]
CO <sub>2</sub> +n-C <sub>7</sub>	310.65-477.21	51	4.2	3.8	1.95	0.73	[17]
CO <sub>2</sub> +n-C <sub>8</sub>	313.15-348.15	20	7.5	5.6	0.92	0.55	[18]
CO <sub>2</sub> +n-C <sub>9</sub>	315.12-418.82	38	4.0	1.5	1.11	0.33	[19]
CO <sub>2</sub> +n-C <sub>10</sub>	462.55-583.65	16	5.9	4.1	1.75	1.23	[20]
CO <sub>2</sub> +n-C <sub>11</sub>	314.98-418.30	42	6.3	1.4	0.13	0.12	[19]
Average			3.9	2.3	1.39	0.91	

**Table 2. Vapor-liquid equilibria of C<sub>1</sub>+alkanes using the SRK equations of state with WS and MWS mixing rules**

System	T [K]	NP	$\Delta P/P$		$\Delta y$		Data ref.
			WS	MWS	WS	MWS	
C <sub>1</sub> +C <sub>2</sub>	210.00-270.00	54	0.9	1.1	0.33	0.37	[12]
C <sub>1</sub> +C <sub>3</sub>	130.37-213.71	105	3.3	2.1	0.10	0.07	[21]
C <sub>1</sub> +n-C <sub>4</sub>	277.59-377.59	24	4.5	1.9	1.25	0.54	[22]
C <sub>1</sub> +n-C <sub>5</sub>	377.59	10	2.5	1.2	0.35	0.33	[23]
C <sub>1</sub> +n-C <sub>7</sub>	199.82-255.37	69	6.7	4.9	0.80	0.63	[24]
C <sub>1</sub> +n-C <sub>8</sub>	223.15-423.15	56	9.4	2.2	0.19	0.13	[25]
C <sub>1</sub> +n-C <sub>9</sub>	223.15-423.15	131	5.1	3.6	0.44	0.40	[26]
C <sub>1</sub> +n-C <sub>10</sub>	542.75-582.15	16	3.9	1.8	0.58	0.23	[27]
Average			4.5	2.3	0.51	0.34	

**Table 3. Vapor-liquid equilibria of C<sub>2</sub>+alkanes using the SRK equations of state with WS and MWS mixing rules**

System	T [K]	NP	$\Delta P/P$		$\Delta y$		Data ref.
			WS	MWS	WS	MWS	
C <sub>2</sub> +C <sub>3</sub>	199.81-255.37	28	2.2	2.2	0.65	0.53	[28]
C <sub>2</sub> +n-C <sub>4</sub>	260.00-280.00	18	4.5	1.1	0.19	0.61	[29]
C <sub>2</sub> +n-C <sub>6</sub>	298.15	7	3.2	2.8	0.55	0.46	[30]
C <sub>2</sub> +n-C <sub>7</sub>	338.70-499.82	32	3.8	1.7	0.98	0.39	[31]
C <sub>2</sub> +n-C <sub>8</sub>	313.15-373.15	46	3.2	2.4	0.94	0.87	[32]
C <sub>2</sub> +n-C <sub>10</sub>	277.60-410.90	66	3.4	2.9	-	-	[33]
Average			3.5	2.2	-	-	

**Table 4. Vapor-liquid equilibria of alcohols+CO<sub>2</sub> using the SRK equations of state with WS and MWS mixing rules**

System	T [K]	NP	$\Delta P/P$		$\Delta y$		Data ref.
			WS	MWS	WS	MWS	
Methanol+CO <sub>2</sub>	313.15-342.80	63	1.7	1.2	0.06	0.04	[34]
Ethanol+CO <sub>2</sub>	313.40-344.75	63	1.5	1.3	0.03	0.03	[34]
1-Butanol+CO <sub>2</sub>	324.16-426.95	23	2.9	2.2	0.21	0.06	[35]
2-Butanol+CO <sub>2</sub>	335.14-431.73	37	4.5	3.6	0.09	0.08	[35]
1-Pentanol+CO <sub>2</sub>	333.08-426.86	38	3.8	2.8	0.03	0.02	[36]
Average			2.9	2.2	0.08	0.05	

where  $\gamma_j^\infty$  is the infinite dilution activity coefficient of species j in i,  $Z_i$  is the pure component liquid phase compressibility at saturation, and  $\phi_j^\infty(T, P_i^{vap})$  is the fugacity coefficient of species j in a mixture at infinite dilution in species i, with  $K_{ji}$  being obtained by index rotation.

## RESULTS AND DISCUSSION

VLE of 30 binary asymmetric systems were investigated. The asymmetric binary systems selected were CO<sub>2</sub>+C<sub>1</sub>-C<sub>11</sub>, C<sub>1</sub>+C<sub>2</sub>-C<sub>10</sub>, C<sub>2</sub>+C<sub>3</sub>-C<sub>10</sub>, and alcohols+CO<sub>2</sub> systems. The modified UNIFAC-

Dortmund model [7,8] was used to calculate the excess Gibbs energy. The interaction parameters required for gas/alkane and alkane/alkane systems were taken from recent studies of the group contribution equation of state based on the UNIFAC model [9-11]. Calculated equilibrium pressures and mole fractions in the vapor phase were compared with the experimental data, and values of the average absolute deviation percent (AAD%) are listed in Tables 1-4.

$$\frac{\Delta P}{P} = \frac{100}{NP} \sum_{i=1}^{NP} \left| \frac{P_i^{exp} - P_i^{calc}}{P_i^{exp}} \right| \quad (12)$$

**Table 5. K<sub>12</sub> and K<sub>21</sub> of some mixtures in the present study**

Binary system	T [K]	K <sub>12</sub>	K <sub>21</sub>
CO <sub>2</sub> -n-C <sub>10</sub>	462.55	0.879	0.475
	476.95	0.892	0.441
	542.95	0.951	0.364
	583.65	0.991	0.388
C <sub>1</sub> -n-C <sub>10</sub>	542.75	0.841	1.319
	563.25	0.839	1.321
	582.15	0.837	1.329
C <sub>2</sub> -n-C <sub>10</sub>	277.60	1.161	1.288
	310.90	1.001	1.755
	344.30	1.062	1.287
	377.60	1.136	1.902
	410.90	1.267	1.593
CO <sub>2</sub> -1Butanol	324.16	0.101	1.513
	333.58	0.112	1.475
	355.38	0.137	1.249
	392.72	0.516	1.100
	426.95	0.631	0.863

**Table 6. Properties of the pure alkanes forming mixtures in the present study [37]**

	Substance	T <sub>c</sub> (K)	P <sub>c</sub> (bar)	$\omega$
1	C <sub>1</sub>	190.7	46.3	0.011
2	CO <sub>2</sub>	304.2	73.9	0.239
3	CH <sub>3</sub> OH	513	80.1	0.556
4	C <sub>2</sub>	305.4	48.8	0.099
5	C <sub>2</sub> H <sub>5</sub> OH	515.8	63.6	0.644
6	C <sub>3</sub>	369.7	42.5	0.153
7	n-C <sub>4</sub>	425.6	37.6	0.199
8	1-C <sub>4</sub> H <sub>9</sub> OH	561.4	44.2	0.592
9	2-C <sub>4</sub> H <sub>9</sub> OH	538	41.8	0.577
10	n-C <sub>5</sub>	470	33.6	0.251
11	1-C <sub>5</sub> H <sub>11</sub> OH	585.9	38.9	0.261
12	n-C <sub>6</sub>	507.9	30.2	0.299
13	n-C <sub>7</sub>	540.1	27.4	0.349
14	n-C <sub>8</sub>	569.1	24.9	0.398
15	n-C <sub>9</sub>	594.8	23	0.445
16	n-C <sub>10</sub>	616.1	21.1	0.489
17	n-C <sub>11</sub>	640.1	19.7	0.535

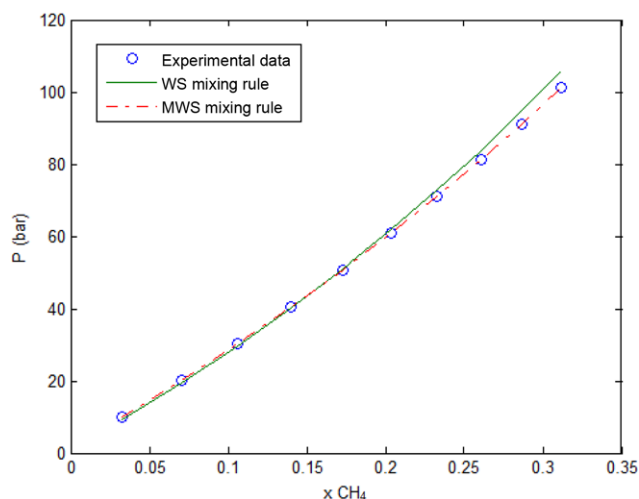


Fig. 1. Experimental data and calculated results of bubble pressure for  $CH_4+n-C_9$  system at 423.15 K.

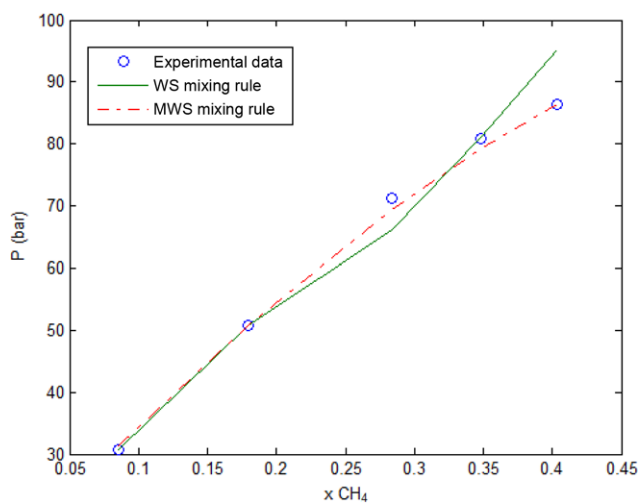


Fig. 2. Experimental data and calculated results of bubble pressure for  $CH_4+n-C_{10}$  system at 583.65 K.

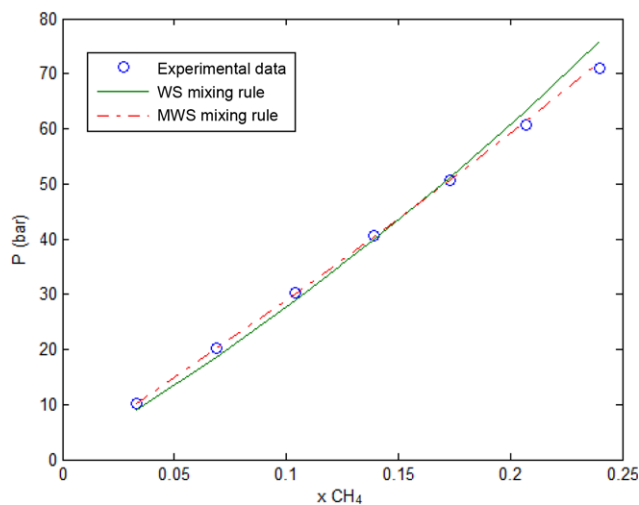


Fig. 3. Experimental data and calculated results of bubble pressure for  $CH_4+n-C_8$  system at 373.15 K.

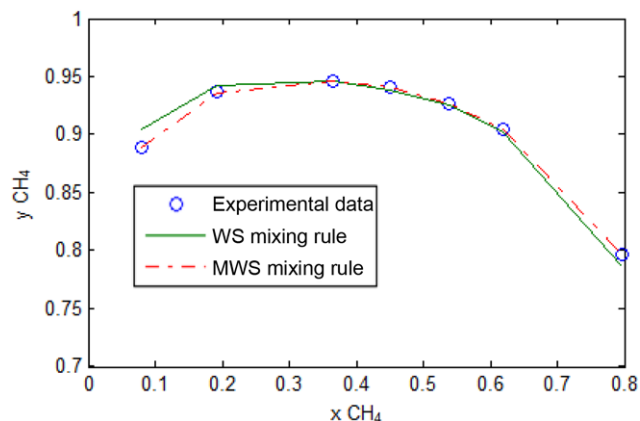


Fig. 4. Experimental data and calculated results of vapor composition for  $CH_4+n-C_4$  system at 277.59 K.

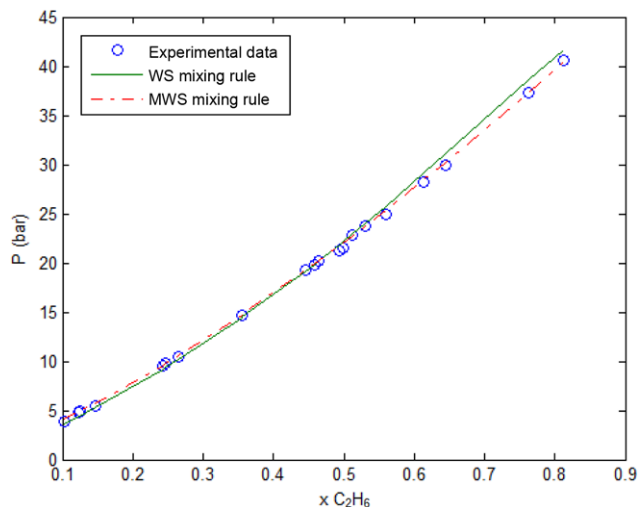


Fig. 5. Experimental data and calculated results of bubble pressure for  $C_2H_6+n-C_{10}$  system at 310.90 K.

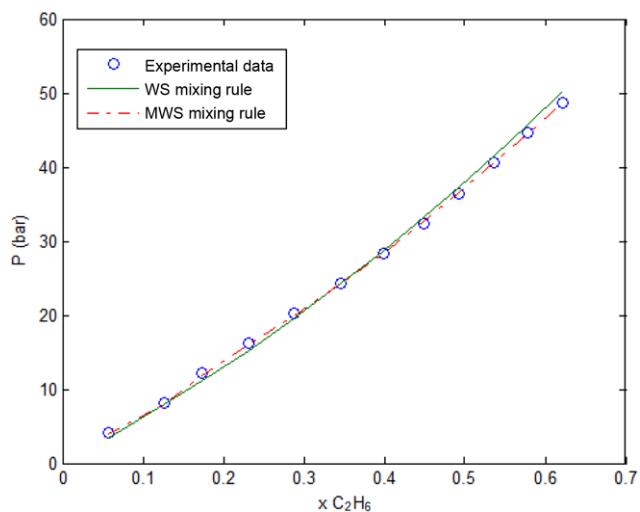


Fig. 6. Experimental data and calculated results of bubble pressure for  $C_2H_6+n-C_8$  system at 348.15 K.

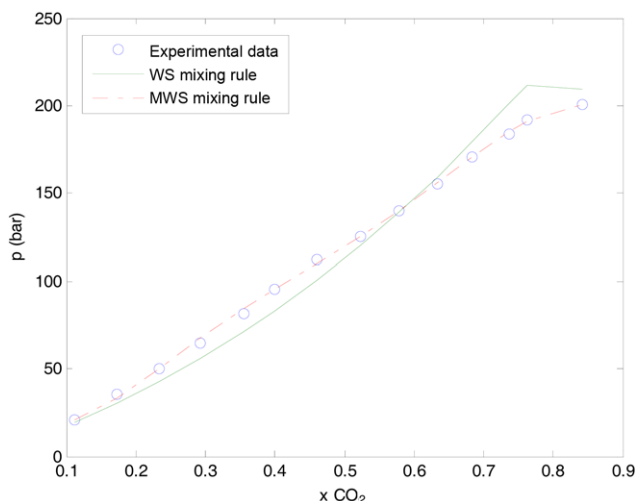


Fig. 7. Experimental data and calculated results of bubble pressure for  $\text{CO}_2 + n\text{-C}_{11}$  system at 418.30 K.

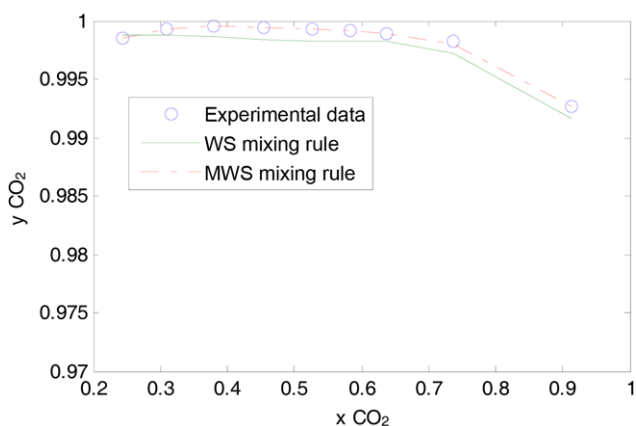


Fig. 8. Experimental data and calculated results of vapor composition for  $\text{CO}_2 + n\text{-C}_{11}$  system at 314.98 K.

$$\Delta y = \frac{100}{\text{NP}} \sum_{i=1}^{\text{NP}} |y_i^{\text{exp}} - y_i^{\text{calc}}| \quad (13)$$

Comparison of the Wong-Sandler and modified Wong-Sandler mixing rules reveals that the results obtained using the modified Wong-Sandler model are more accurate.

The two parameters  $K_{12}$  and  $K_{21}$  included in our model are reported for some systems in Table 5. The properties of the pure substances included in the mixtures studied are given in Table 6.

Figs. 1 through 9 depict the comparison between experimental data and predicted results using WS and MWS model for some systems. The results demonstrate that the MWS model is more accurate and reliable.

## CONCLUSION

The proposed model in this work was based on a modification of Wong-Sandler mixing rule to study asymmetric mixtures consisting of light components and heavy hydrocarbons. In this model, a composition-dependent binary interaction parameter ( $k_{ij}$ ) was used to improve the ability of the Wong-Sandler mixing rule for the pre-

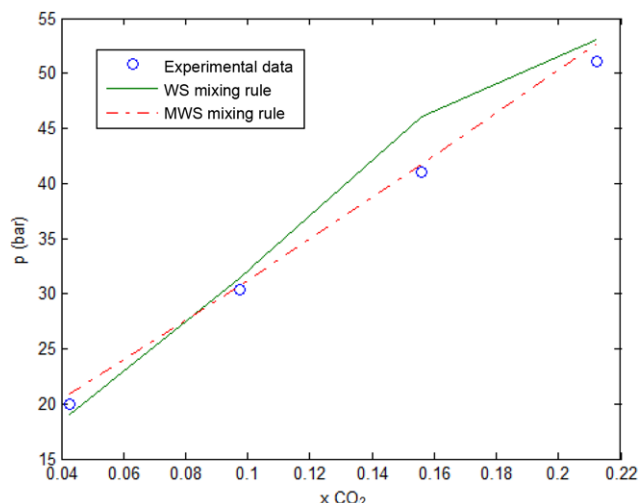


Fig. 9. Experimental data and calculated results of bubble pressure for  $\text{CO}_2 + n\text{-C}_{10}$  system at 583.65 K.

diction of vapor-liquid equilibria in asymmetric systems. VLE of 30 binary systems were investigated and the comparison between Wong-Sandler and modified Wong-Sandler mixing rules revealed that the results obtained using the proposed model were more accurate and reliable.

## NOMENCLATURE

### List of Symbols

- $a$  : attraction parameter in equation of state
- $\underline{A}^E$  : molar excess Helmholtz free energy
- $b$  : co volume parameter in equation of state
- $C$  : equation of state dependent constant in Wong-Sandler mixing rule
- $\underline{G}^E$  : molar excess Gibbs free energy
- $k_{ij}$  : binary interaction parameter
- $K_{ij}, K_{ji}$  : two parameters defined in Eq. (7)
- NP : number of data
- $P$  : pressure
- $P_c$  : critical pressure
- $R$  : universal gas constant
- $T$  : temperature
- $T_c$  : critical temperature
- $u, w$  : constant of equation of state
- $\underline{V}$  : molar volume
- $x$  : liquid mole fraction
- $y$  : vapor mole fraction

### Greek Symbols

- $\phi$  : fugacity coefficient
- $\gamma$  : activity coefficient
- $\omega$  : acentric factor

### Abbreviations

- EOS : equation of state
- MWS : modified Wong-Sandler mixing rule
- SRK : Soave-Redlich-Kwong equation of state

VLE : vapor-liquid equilibrium  
 WS : Wong-Sandler mixing rule

### Subscript/Superscript

exp : experimental  
 i, j : component in a mixture  
 m : mixture

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