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Short communication

Correlation of vapor–liquid equilibrium for systems of carbon dioxide + hydrocarbon by the corresponding-states principle

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Abstract

A combination of the corresponding-states principle with mixing rules is used to correlate vapor–liquid equilibrium data for systems of carbon dioxide + hydrocarbon. Good agreement with experimental data is obtained by a suitable choice of the equations of state up to medium asymmetric systems. Binary interaction parameters for the proposed method are given. © Elsevier Science S.A.

Keywords: Equation of state; Thermodynamics; Vapour–liquid equilibria; Hydrocarbons; Binary mixtures; Symmetric mixtures

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1. Introduction

Fluid phase equilibrium data are essential elements in the design and development of industrial processes. The phase behavior of carbon dioxide with hydrocarbon is of interest in the processing of petroleum products, the production of coal liquids and enhanced oil recovery operations.

Prediction of vapor–liquid equilibria of asymmetric mixtures by use of the corresponding-states principle has been developed by many researchers. Joffe [1] and Plöcker et al. [2] have used the Lee and Kesler correlation [3] with the pseudocritical method. Arai et al. [4] and Tsai and Shyu [5] presented the mixing rules combining functionally different equations of state on the basis of the three-parameter corresponding-states principle. Wong et al. [6,7] used a generalized corresponding-states principle based on the properties of two nonspherical reference fluids for the calculation of vapor–liquid equilibria. Mentzer et al. [8] and Al-Sahhaf et al. [9] used the principle of corresponding states to calculate vapor–liquid equilibria for systems of light gases with light hydrocarbons.

In this work we use a one-fluid theory with mixing rules on the basis of the corresponding-states principle to calculate the vapor–liquid equilibria for systems of carbon dioxide with hydrocarbons. Data used in evaluating this method are taken from the literature.

2. Corresponding-states principle with mixing rules

The basic thermodynamic requirement for a vapor–liquid equilibrium is

$$f_i^{\mathrm{V}} = f_i^{\mathrm{L}} \tag{1}$$

Replacing fugacities with fugacity coefficients, Eq. (1) becomes

$$y_i \phi_i^{\mathrm{V}} = x_i \phi_i^{\mathrm{L}} \tag{2}$$

Following Van Ness [10], the fugacity coefficient of component i in the mixture at constant T and P is related to the mixture fugacity coefficient by

$$\begin{aligned}\ln \phi_i = \ln \phi_M &- \frac{1}{RT}\frac{\Delta h_{\mathrm{M}}}{T_{\mathrm{CM}}}\sum_{j\neq i} z_j\left(\frac{\mathrm{d}T_{\mathrm{CM}}}{\mathrm{d}z_j}\right)_{zk} \\ &+ \frac{Z_{\mathrm{M}}-1}{P_{\mathrm{CM}}}\sum_{j\neq i} z_j\left(\frac{\mathrm{d}P_{\mathrm{CM}}}{\mathrm{d}z_j}\right)_{zk} \\ &- \left(\frac{\partial \ln \phi_M}{\partial \omega_{\mathrm{M}}}\right)_{T_{\mathrm{r}},P_{\mathrm{r}}}\sum_{j\neq i} z_j\left(\frac{\mathrm{d}\omega_{\mathrm{M}}}{\mathrm{d}z_j}\right)_{zk} \quad k\neq i,j\end{aligned} \tag{3}$$

where $\ln \phi_{\mathrm{M}}$, $(\Delta h/RT_{\mathrm{C}})_{\mathrm{M}}$ and Z_{M} are obtained from the Teja correlation [11–14]:

$$g_{\mathrm{M}}(T_{\mathrm{r}},P_{\mathrm{r}},\omega_{\mathrm{M}}) = \sum_i z_i g^{(i)}(T_{\mathrm{r}},P_{\mathrm{r}},\omega_i) \tag{4}$$

To calculate Z_{M}, $\ln \phi_{\mathrm{M}}$ and $(\Delta h/RT_{\mathrm{C}})_{\mathrm{M}}$ we use a one-fluid theory. The pseudocritical method, similar to that of Plöcker

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Table 1
Comparison between calculated and experimental vapor–liquid equilibria for nineteen carbon dioxide (1) + hydrocarbon (2) systems

Component 2	Temperature (K)	Optimum k_{ij}			DEVP (%)			DELY × 10^2			Ref.
		SRK	PR	PT	SRK	PR	PT	SRK	PR	PT	
CH_4	193.2	0.969	0.965	0.961	0.18	0.13	0.40	0.17	0.15	0.16	[23]
	208.5	0.971	0.971	0.972	0.41	0.41	0.45	1.04	1.04	1.06	[24]
	230.0	0.992	0.992	0.987	1.63	0.77	1.74	1.22	1.22	0.71	[25]
	250.0	0.999	1.000	1.001	0.34	0.42	0.54	1.32	1.26	1.57	[25]
	270.0	0.995	0.994	1.002	1.11	1.05	1.39	0.90	0.58	1.00	[25]
C_2H_4	231.6	0.963	0.961	0.959	0.99	0.80	1.42	1.37	1.26	1.48	[26]
	253.0	0.964	0.964	0.959	0.72	0.72	1.73	0.33	0.33	0.49	[26]
	273.2	0.951	0.950	0.961	5.56	5.56	1.06	3.61	3.62	0.88	[27]
C_2H_6	222.0	0.900	0.899	0.900	0.47	0.53	0.47	1.78	1.85	1.67	[28]
	250.0	0.906	0.905	0.906	0.17	0.19	0.14	0.46	0.51	1.01	[25]
	273.0	0.899	0.900	0.899	1.56	1.73	1.78	0.54	0.52	0.58	[26]
	288.2	0.909	0.907	0.911	0.51	0.26	0.82	0.80	0.57	0.77	[29]
	298.2	0.913	0.912	0.913	0.04	0.06	0.04	0.76	0.68	0.77	[29]
C_3H_6	253.0	0.964	0.963	0.961	3.07	3.06	3.09	0.59	0.58	0.59	[30]
	273.2	0.939	0.939	0.938	1.75	1.75	1.65	1.14	1.13	1.12	[30]
C_3H_8	244.1	0.922	0.922	0.918	2.34	2.35	2.30	0.47	0.47	0.53	[31]
	277.4	0.927	0.928	0.923	1.59	1.59	1.38	0.53	0.47	0.66	[32]
	310.8	0.924	0.924	0.927	1.57	1.57	1.52	0.78	0.78	0.73	[32]
	327.4	0.928	0.927	0.930	0.80	0.79	0.93	0.53	0.51	0.72	[32]
	344.1	0.930	0.930	0.932	0.48	0.48	0.88	1.21	1.21	3.10	[32]
1-C_4H_8	273.2	0.991	0.992	0.988	3.00	3.00	2.91	0.67	0.66	0.63	[30]
n-C_4H_{10}	227.8	0.939	0.934	0.936	2.20	1.24	1.82	0.70	0.60	0.60	[33]
	283.0	0.957	0.954	0.953	2.69	2.13	2.48	0.48	0.40	0.48	[33]
	344.1	0.964	0.971	0.954	4.08	2.02	4.15	1.28	0.92	1.51	[34]
	377.4	0.969	0.969	0.958	1.16	1.70	1.98	0.57	1.06	0.93	[34]
	410.8	0.969	0.969	0.954	0.74	0.57	0.15	1.49	1.52	1.38	[34]
iso-C_4H_{10}	273.2	0.948	0.945	0.944	4.98	5.00	5.12	1.46	1.47	1.46	[26]
	310.9	0.954	0.956	0.951	2.40	2.21	2.13	1.63	1.58	1.25	[35]
	344.3	0.955	0.960	0.952	3.20	3.20	2.83	0.56	0.61	0.44	[35]
	377.6	0.957	0.959	0.957	1.87	2.00	1.92	1.15	1.22	0.87	[35]
	394.3	0.954	0.951	0.950	1.27	1.29	0.46	0.79	0.62	1.37	[35]
n-C_5H_{12}	277.7	0.995	0.994	0.990	2.53	2.57	2.60	0.40	0.40	0.39	[36]
	311.0	1.003	1.003	0.999	3.78	3.79	3.13	0.31	0.31	2.29	[36]
	344.2	1.014	1.013	1.009	4.03	3.76	3.70	2.02	3.19	2.16	[36]
	377.6	1.015	1.015	1.009	2.93	2.93	2.80	1.44	1.44	1.23	[36]
iso-C_5H_{12}	277.6	0.991	0.991	0.988	3.02	3.03	2.78	0.37	0.38	0.32	[37]
	310.9	0.999	0.999	0.994	4.02	4.04	3.92	0.37	0.37	0.35	[37]
	344.3	1.001	1.001	0.997	3.27	3.25	3.05	1.09	1.09	1.13	[37]
	377.7	1.004	1.005	1.000	2.31	2.29	1.91	1.22	1.22	1.23	[37]
C_6H_6	298.2	1.029	1.020	1.022	2.72	2.62	2.66	0.15	0.11	0.11	[38]
	313.2	1.033	1.026	1.027	2.67	2.15	2.31	0.42	0.37	0.38	[38]
C_6H_{12}	473.2	1.085	1.081	1.082	0.95	0.79	0.90	1.75	2.05	1.95	[39]
	483.2	1.063	1.070	1.070	0.77	1.05	1.19	1.99	2.33	2.37	[39]
	503.2	1.045	1.054	1.052	0.73	0.91	0.97	2.84	2.83	2.90	[39]
	523.2	1.077	1.075	1.064	0.37	0.52	1.43	1.79	1.54	3.05	[39]
	533.2	1.095	1.095	1.095	0.16	0.13	0.10	1.03	0.99	0.95	[39]
n-C_6H_{14}	298.0	1.042	1.042	1.036	4.27	4.23	4.02	0.50	0.49	0.49	[38]
	313.0	1.053	1.053	1.039	3.40	3.34	5.71	0.39	0.39	0.50	[38]
	353.2	1.045	1.045	1.042	5.67	5.63	5.26	1.31	1.29	1.21	[40]
	393.2	1.055	1.056	1.043	3.56	3.57	5.23	1.33	1.33	1.67	[40]
$C_6H_5CH_3$	311.3	1.059	1.051	1.052	1.84	1.41	1.39	0.15	0.17	0.16	[41]
	352.6	1.066	1.060	1.060	4.60	3.76	3.94	0.86	0.78	0.79	[41]
	393.7	1.083	1.077	1.079	7.03	6.20	6.26	1.65	1.60	1.62	[41]
	477.0	1.091	1.089	1.089	2.75	2.44	2.48	2.68	2.87	2.87	[41]
n-C_7H_{16}	310.7	1.097	1.097	1.091	7.69	7.69	7.00	0.22	0.22	0.21	[41]
	352.6	1.101	1.101	1.098	7.42	7.42	6.62	0.71	0.71	0.71	[41]
	394.3	1.116	1.116	1.123	5.00	5.00	2.14	1.42	1.42	1.19	[41]
	477.2	1.159	1.148	1.146	633	1.12	1.25	7.87	5.71	4.74	[41]

(*continued*)

Table 1 (*continued*)

Component 2	Temperature (K)	Optimum k_{ij}			DEVP (%)			DELY × 10^2			Ref.
		SRK	PR	PT	SRK	PR	PT	SRK	PR	PT	
$C_{10}H_{14}$	273.2	1.146	1.146	1.134	6.80	6.76	5.21	0.01	0.28	0.01	[42]
	293.2	1.154	1.155	1.143	5.70	5.71	4.16	0.01	1.26	0.01	[42]
n-$C_{10}H_{22}$	277.6	1.189	1.177	1.176	12.78	11.94	11.62	0.00	0.00	0.00	[43]
	377.6	1.220	1.214	1.216	8.80	8.25	7.49	1.55	1.41	1.44	[43]
	410.9	1.231	1.227	1.229	7.79	7.33	6.54	1.51	1.35	0.22	[43]
	444.3	1.240	1.237	1.237	6.27	4.94	3.92	0.34	0.31	0.26	[43]
	510.9	1.250	1.249	1.251	3.63	2.75	2.19	0.85	1.06	1.23	[43]
n-$C_{16}H_{34}$	463.1	1.462	1.462	1.457	0.44	0.44	2.47	0.06	0.06	0.07	[44]
	542.6	1.478	1.477	1.479	1.21	1.22	0.42	0.38	0.38	0.10	[44]
	623.6	1.494	1.494	1.502	0.53	0.53	1.13	0.17	0.17	1.12	[44]
	663.8	1.507	1.508	1.514	0.40	0.40	1.06	1.44	1.42	3.54	[44]
n-$C_{20}H_{42}$	310.2	1.522	1.522	1.499	18.77	18.79	16.64	0.00	0.00	0.00	[45]
	323.2	1.530	1.530	1.506	10.96	10.53	10.27	0.01	0.01	0.00	[45]
	348.2	1.538	1.538	1.518	4.75	4.76	5.62	0.04	0.04	0.00	[45]
	373.2	1.540	1.540	1.519	1.09	1.12	2.61	0.17	0.02	0.00	[45]
Overall					3.35	3.14	3.02	0.95	0.96	0.96	

$$\mathrm{DEVP}=\frac{1}{n}\sum_i^n\left|\frac{P_{\mathrm{cal}}-P_{\mathrm{exp}}}{P_{\mathrm{exp}}}\right|_i\times 100\%$$

$$\mathrm{DELY}=\frac{1}{n}\sum_i^n|y_{\mathrm{cal}}-y_{\mathrm{exp}}|_i$$

et al. [2], is used here, and a set of mixing rules for the averaging procedure to calculate the pseudocritical constants of a mixture is proposed as follows:

$$T_{\mathrm{CM}}V_{\mathrm{CM}}^{\eta}=\sum_i\sum_j z_iz_jT_{cij}V_{cij}^{\eta} \tag{5}$$

$$V_{\mathrm{CM}}=\sum_i\sum_j z_iz_jV_{cij} \tag{6}$$

$$T_{cij}=k_{ij}(T_{ci}T_{cj})^{1/2} \tag{7}$$

$$V_{cij}=\tfrac{1}{8}(V_{ci}^{1/3}+V_{cj}^{1/3})^3 \tag{8}$$

$$P_{\mathrm{CM}}=Z_{\mathrm{CM}}RT_{\mathrm{CM}}/V_{\mathrm{CM}} \tag{9}$$

$$Z_{\mathrm{CM}}=0.2905-0.085\omega_{\mathrm{M}} \tag{10}$$

$$\omega_{\mathrm{M}}=\sum_i Z_i\omega_i \tag{11}$$

3. Calculation procedure

The calculation procedure for vapor–liquid equilibria is similar to that of Plöcker et al. [2] and Arai et al. [4]. The equations of state used for the pure components are written in reduced form using the following equations:

$$T_{\mathrm{r}}=\frac{T}{T_{\mathrm{c}}},\ P_{\mathrm{r}}=\frac{P}{P_{\mathrm{c}}},\ V_{\mathrm{r}}=\frac{V}{V_{\mathrm{c}}},\ Z=\frac{P_{\mathrm{r}}V_{\mathrm{r}}}{T_{\mathrm{r}}}$$

For the calculated values of $T_{\mathrm{r}}(=T/T_{\mathrm{CM}})$ and $P_{\mathrm{r}}(=P/P_{\mathrm{CM}})$ the reduced volumes can be found from the reduced equation of state. The largest root obtained, $V_{\mathrm{r}}^{(i),\mathrm{V}}$, is used to evaluate the vapor-phase properties, while the smallest root obtained, $V_{\mathrm{r}}^{(i),\mathrm{L}}$, is used to evaluate the liquid-phase properties. Then Z_{M}, ln ϕ_{M} and $(\Delta h/RT_{\mathrm{C}})_{\mathrm{M}}$ are obtained from Eq. (4), and Eqs. (A3)–(A5) for vapor and liquid phases. Finally the vapor–liquid equilibrium calculations are performed by using Eqs. (2)–(11), and Eqs. (A1)–(A7).

4. Results and discussion

The mixing rules proposed as shown in Eqs. (5)–(11) are similar to those proposed previously by many researchers [2,4,19–22]. If we set $\eta=0$ we obtain mixing rules as used by Prausnitz and Gunn [15] and Barner and Quinlan [16]. If we set $\eta=1$ we obtain mixing rules similar to those of Leland et al. [17], Mollerup [18] and Arai et al. [4]. Plöcker et al. [2] used the Lee-Kesler correlation [3] to calculate vapor–liquid equilibria and they found that good agreement with experiment is obtained by setting $\eta=0.25$.

In this study, bubble pressure calculations are used to correlate the vapor–liquid equilibria for systems of CO_2 + hydrocarbon. The equation of state proposed by Huang et al. [19] is used to describe the P–V–T relation for carbon dioxide, while for the hydrocarbons the Soave–Redlich–Kwong equation [20], the Peng–Robinson equation [21] or the Patel–Teja equation [22] is used. For each calculation with a given value of η, an optimal binary parameter k_{12} is obtained by minimizing the bubble pressure variance. It can be seen that the exponent η appearing in Eq. (5) is of no practical importance for slightly asymmetric mixtures, but it becomes significant for very asymmetric mixtures. We have determined η values by a systematic study of numerous asymmetric mixtures. The calculated results show that the best values of η change somewhat from one system to another, and good agreement with experimental data is obtained by

Table 2
Average DEVP and DELY for nineteen carbon dioxide (1) + hydrocarbon (2) systems

Component 2	Temperature range (K)	Max. P (atm)	Data points	DEVP (%)			DELY × 10^2		
				SRK	PR	PT	SRK	PR	PT
CH_4	193.2–270.0	84.1	52	0.73	0.55	0.90	0.93	0.85	0.90
C_2H_4	231.6–273.2	43.4	36	2.42	2.36	1.41	1.77	1.74	0.95
C_2H_6	222.0–298.2	65.4	61	0.55	0.56	0.65	0.87	0.82	0.96
C_3H_6	253.0–273.2	34.4	27	2.41	2.40	2.37	0.86	0.86	0.86
C_3H_8	244.1–344.1	68.0	65	1.36	1.36	1.40	0.70	0.69	1.15
1-C_4H_8	273.2	34.4	13	3.00	3.00	2.91	0.67	0.66	0.63
n-C_4H_{10}	277.8–410.8	78.2	54	2.17	1.53	2.12	0.90	0.90	0.98
iso-C_4H_{10}	273.2–394.3	69.9	51	2.74	2.74	2.49	1.12	1.10	1.08
n-C_5H_{12}	277.7–377.6	95.1	53	3.32	3.25	3.06	1.04	1.33	1.52
iso-C_5H_{12}	277.6–377.7	87.8	46	3.16	3.15	2.92	0.76	0.76	0.76
C_6H_6	298.2–313.2	76.5	17	2.70	2.38	2.48	0.29	0.24	0.25
C_6H_{12}	473.2–533.2	100.0	29	0.60	0.68	0.92	1.88	1.95	2.24
n-C_6H_{14}	298.0–393.2	114.4	49	4.22	4.19	5.05	0.88	0.88	0.97
$C_6H_5CH_3$	311.3–477.0	150.9	34	4.05	3.45	3.52	1.34	1.35	1.36
n-C_7H_{16}	310.7–477.2	131.4	62	6.61	5.31	4.25	2.55	2.01	1.71
$C_{10}H_{14}$	273.2–293.2	50.0	14	6.25	6.23	4.69	0.01	0.77	0.01
n-$C_{10}H_{22}$	277.6–510.9	170.1	57	7.86	7.04	6.35	0.85	0.83	0.63
n-$C_{16}H_{34}$	463.1–663.8	50.2	16	0.64	0.65	1.27	0.51	0.51	1.21
n-$C_{20}H_{42}$	310.2–373.2	75.0	60	8.89	8.80	8.78	0.05	0.02	0.00
Overall			796	3.35	3.14	3.02	0.95	0.96	0.96

setting $\eta = 0.35$ for all binary systems studied in this work. This value of η is the same as in our previous work [5].

The calculated best values of binary parameter k_{ij} at various temperatures and average DEVP and DELY for 19 carbon dioxide + hydrocarbon systems are listed in Tables 1 and 2. From Table 1 binary parameters are found to be slightly dependent on temperature. Table 2 compares the deviations in calculated and experimental bubble pressure and vapor composition between those obtained by the SRK, PR and PT equations of state. It can be seen that the calculated results do not differ appreciably when using different equations of state for hydrocarbons.

5. Conclusion

The vapor–liquid equilibria for systems of carbon dioxide with hydrocarbons at temperatures ranging from 193 to 511 K and pressures up to 170 atm were successfully correlated by using the corresponding states principle in combination with classical mixing rules using optimum binary interaction parameters k_{ij} and universal constant $\eta = 0.35$. In general the calculated results are in good agreement with experimental data for symmetric or slightly asymmetric mixtures possibly up to n-C_{16}/CO_2, whereas they are in poor agreement with experimental data for very asymmetric mixtures.

6. Nomenclature

f	fugacity (atm)
g	thermodynamic property
Δh	molar residual enthalpy (atm cm^3 mol^{-1})
k_{ij}	binary parameter for the i–j mixture
P	pressure (atm)
R	gas constant (atm cm^3 mol^{-1} K^{-1})
T	temperature (K)
V	molar volume (cm^3 mol^{-1})
x	mole fraction in liquid phase
y	mole fraction in vapor phase
z	mole fraction (liquid or vapor phase)
Z	compressibility factor

6.1. Greek letters

η	universal exponent in the mixing rule for the pseudocritical temperature
ϕ	fugacity coefficient
ω	acentric factor

6.2. Subscripts

CM	pseudocritical property
C	critical property
M	mixture property
i, j, k	components i, j and k
r_1, r_2	reference fluid
r	reduced property

6.3. Superscripts

L	liquid phase
V	vapor phase
(r), (r_1), (r_2)	reference fluid

(o) simple fluid
(1), (2), (*i*), (*n*) component 1, 2, *i* and *n*

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Appendix A

The expression for the derivatives $(\partial \ln \phi_M/\partial\omega_M)_{T_r,P_r}$ and $(d\omega_M/dz_j)_{z_k}$ in Eq. (3) depends on the form of the three-parameter corresponding states principle used. Following Eqs. (4) and (11) we have

$$\left(\frac{\partial \ln \phi_M}{\partial \omega_M}\right)_{T_r,P_r} = \frac{\ln \phi_M - \ln \phi^{(i)}}{\omega_M - \omega_i} \qquad \text{(A1)}$$

and

$$\left(\frac{d\omega_M}{dz_j}\right)_{z_k} = \omega_j - \omega_i \qquad \text{(A2)}$$

The three quantities Z_M, $\ln \phi_M$ and $(\Delta h/RT_C)_M$ in Eq. (3) can be obtained by combining Eq. (4) with the following equations:

$$Z^{(i)} = \frac{P_r V_r^{(i)}}{T_r} \qquad \text{(A3)}$$

$$\ln \phi^{(i)} = -\frac{1}{T_r}\int_{\infty}^{V_r^{(i)}} \left(P_r - \frac{T_r}{V_r}\right) dV_r + Z^{(i)} - 1 - \ln Z^{(i)} \qquad \text{(A4)}$$

$$\left(\frac{\Delta h}{RT_C}\right)^{(i)} = \int_{\infty}^{V_r^{(i)}} \left[T_r\left(\frac{\partial P_r}{\partial T_r}\right)_{V_r} - P_r\right] dV_r + T_r(Z^{(i)} - 1) \qquad \text{(A5)}$$

The derivatives of the pseudocriticals in Eq. (3) can be obtained from the above mixing rules as follows:

$$\left(\frac{dT_{CM}}{dz_j}\right)_{z_k} = (2\sum_l z_l(T_{clj}V_{cil}^{\eta} - T_{cli}V_{clj}^{\eta}) - 2\eta V_{CM}^{\eta-1}\sum_l z_l(V_{clj} - V_{cli})T_{CM})/V_{CM}^{\eta} \qquad \text{(A6)}$$

$$\left(\frac{dP_{CM}}{dz_j}\right)_{z_k} = P_{CM}(-0.085(\omega_j - \omega_i)/V_{CM} - 2\sum_l z_l(V_{clj} - V_{cli})/V_{CM} + (dT_{CM}/dz_j)_{z_k}/T_{CM}) \qquad \text{(A7)}$$

In Eqs. (A6) and (A7), $k \neq i,j$.

References

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