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Modification of Peng-Robinson Equation of State for Supercritical Carbon Dioxide

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Abstract

Use of carbon dioxide in supercritical process is increasingly interested, especially in power cycles and refrigeration systems. To find the thermodynamic properties for system design and analysis, the accurate equation of state is required. In the study, the parameter a in Peng-Robinson equation of state was modified for supercritical carbon dioxide. Two forms of new parameter a were proposed and the coefficients in the parameter were found based on the $P\rho T$ data obtained from Span and Wagner equation of state. The predicted density from both modified equations was compared with original Peng-Robinson and Span and Wagner equations of state. Moreover, the density at pressure and temperature outside fitting range was also investigated the accuracy. The results showed that both forms of modified Peng-Robinson equation of state can provide fairly better predicted result than that from original Peng-Robinson equation of state.

Keywords: Alpha function, Peng-Robinson Equation of State, Supercritical CO_2

1. INTRODUCTION

Combustion of fossil fuel is a major source of carbon dioxide (CO_2) emission which causes greenhouse effect. Carbon capture and sequestration (CCS) is a method that CO_2 is captured at the source and transported to a storage site, which usually is an underground geological formation. Another attractive way is use of CO_2 as a raw material for producing useful gas [1,2] or as working fluid in power cycle as well as refrigerant [3-9]. Thus, CO_2 has become an important substance in many applications, especially in high pressure and temperature, called supercritical range (P>73.8 bar, T>304.13K) [3-9]. For design and analysis of system using CO_2 as working fluid, thermodynamic properties are required. Usually, these properties, for examples: enthalpy, internal energy, and density, are obtained from the calculation, in which equation of state is needed. An accurate equation of state for CO_2 has been developed by Span and Wagner (SW) [10]. The detail of SW equation is introduced in Appendix. Heidaryan and Jarrahian [11] informed that the Span and Wagner (SW) equation of state is considered as the top choice of equation of state for predicting the properties of CO_2. However, they gave the opinion that SW equation of state is over-complicated and time consuming for computer calculation. Kim [12] has mentioned that SW equation of state is accurate but it contains many terms. Some of the terms are complex exponential which become difficult for computation. Recently, Aavatsmark et al. [13] have also confirmed that SW equation of state consisting of nonlinear terms that are expensive to solve with the numerical solution.

Cubic equation of state is often used for prediction of thermodynamic properties. It has been well-accepted that Peng-Robinson (PR) and Soave-Redlich-Kwong (SRW) equations of state are the most popular equations of state [14]. Their modifications to improve the accuracy of the property prediction may be categorized in two approaches [11]. The first method is development of a new attractive or repulsive terms and the second approach is improvement of temperature dependence of the attractive parameter through a new alpha function. Gasem et al. [15] have developed a new alpha function in PR equation of state for different molecular species, including heavy hydrocarbons. Coquelet et al. [16] investigated a new form of alpha function combining both exponential and polynomial forms. The prediction results of pure-compound vapor pressures from use of the new alpha function significantly improved. The modification of alpha function for the PR equation of state has been found in Ref [14] for non-hydrocarbon. A new alpha function, which was represented in exponential form, was study again by Saffari and Zahedi [17]. It was applied to predict the vapor-pressure of natural gas. The new alpha functions mentioned above were mostly developed for vapor pressure prediction. It means that subcritical region was often considered. However, some researches were focused on thermodynamic property prediction in supercritical region. Skamenca and Tassios [18] found two new constants (a and b) in Redlich-Kwong equation of state by using regression method. The study ranges were cover subcritical and supercritical regions. The behavior of alpha function, also called Soave function, for various gases at supercritical temperature was investigated by Mahmoodi and Sedigh [19]. For developed new alpha function, Heidary and Jarrahian [11] have modified alpha function in Redlich-Kwong equation of state for supercritical CO_2. The new alpha function was presented as β function which was a function of natural logarithm of reduced temperature, $\ln(T_r)$, and natural logarithm of reduced pressure, $\ln(P_r)$.

From details mentioned above, it can be observed that a few researches have been done for supercritical region. Moreover, the β function in Ref [11] used six coefficients and its form is quite complicated. In this research, the parameter a in PR equation of state is modified for CO_2 in supercritical region. The same form of parameter a as PR equation of state is used and the constants are found matching with the data in supercritical region, obtained from SW equation of state [11]. Another modification is that the parameter a is presented in exponential form and the constants are calculated. The densities predicted by the modified PR equations of state are compared with that obtained from the SW and the original PR equations of state.

2. METHOD

2.1 Modification of equation of state

Cubic equation of state, such as the van der Waals (vW), Redlich-Kwong (RW), SRK and PR equations of state, can be written in the form [20]

$$P=\frac{RT}{v-b}-\frac{a}{(v+c)(v+d)} \tag{1}$$

Where b , c, and d are constants, and a is a function of temperature. Table 1 shows the constants for above mentioned cubic equations of state.

For Sove and PR equations of state, the alpha function, α , in parameter a first proposed by Soave [21] and it is defined as:

$$\alpha=\left[1+\kappa\left(1-\sqrt{T_r}\right)\right]^2 \tag{2}$$

where κ is a function of ω called the acentric factor. For PR equation of state, the parameter κ is given by

$$\kappa=0.37464+1.54226\omega-0.26992\omega^2 \tag{3}$$

In this study, the parameter a for PR equation of state is modified for supercritical CO_2. The modified of parameter a in PR equation is given by

$$a_{MPR1}=k_1\frac{R^2T_c^2}{P_c}\left[1+k_2\left(1-\sqrt{T_r}\right)\right]^2 \tag{4}$$

The first modification is done for a_c term. The constant is changed to be k_1 which is fitted with the data of supercritical CO_2. The second change is conducted in α function. The parameter κ is determined its value associated with PvT (or $P\rho T$) data in supercritical region and it is set as k_2. Moreover, the exponential form of alpha function, proposed by Trebble and Bishnoi [22] is also investigated in this study. This modification of parameter a is defined as

Table 1 Constants in Eq. (1) for cubic equations of state

Equation of state	***c***	***d***	***b***	***a***
vW	0	0	$\frac{RT_c}{8P_c}$	$\frac{27}{64}\frac{R^2T_c^2}{P_c}$
RK	b	0	$\frac{0.08664RT_c}{P_c}$	$\frac{0.42748RT_c^{2.5}}{P_cT^{0.5}}$
SRK	b	0	$\frac{0.08664RT_c}{P_c}$	$\frac{0.42748R^2T_c^2}{P_c}\alpha=a_c\alpha$
PR	$\left(1+\sqrt{2}\right)b$	$\left(1-\sqrt{2}\right)b$	$\frac{0.07780RT_c}{P_c}$	$\frac{0.45724R^2T_c^2}{P_c}\alpha=a_c\alpha$

$$a_{MPR2}=k_3\frac{R^2T_c^2}{P_c}\exp\left[k_4\left(1-T_r\right)\right] \tag{5}$$

This form of alpha function has been cited in Ref [19] that Mathias and Copenman, [23] used this alpha function form for highly polar components in supercritical region, $(T_r>1)$ and it provided good predicted result. Thus, the exponential form is applied in this study. It is worth to discuss here that both forms of alpha function are simple and consist only of two constants for each function. For calculating departure functions, for example: enthalpy departure, the complicated form of alpha function may lead to cumbersome process to find those functions, especially differentiation of parameter a .

2.2 Determination of coefficients

As mentioned in introduction part, SW equation of state has been particularly developed for CO_2 with high accurate property prediction. Therefore, SW equation of state is selected to generate the PρT data used to determine the values of coefficients k_1 to k_4 . At certain P and T, a density of CO_2 is found by SW equation of state. The density is, then, substitutes into Eq. (1) to find the value of parameter a and it is assigned as a_{MPR}. Optimization method, called Levenberg-Marquardt (LM) [24], is applied to find k_1 and k_2 in Eq. (4). For LM method, it is explained in the next section. To find coefficients k_3 and k_4 in Eq. (5), the same procedure, mentioned above, is repeated.

In this study, the range of pressure and temperature focused to find the new coefficients are 75 bar to 147.6 bar and 306 K to 700 K, respectively, which covers the application rages in Ref. [3-9]. However, the modified equation of state, obtained from data fitting, will be also tested at outside fitting range.

2.3 Levernberg-Marquardt

For least square curve fitting problem, there are data (x_i, y_i) which are required to model. A model function $f\left(x_i,k\right)$ is defined and k is found the value that causes minimum sum of the squares of the deviations between y_i and $f\left(x_i,k\right)$, called r :

$$r=\sum_1^M\left(y_i-f\left(x_i,k\right)\right)^2=\sum_1^M\left(\varepsilon_i\right)^2 \tag{6}$$

In Levernberg Marquardt algorithm, the initial value of k is defined. The iterative procedure is carried out until the parameter k converges the solution. For the first iteration, the value of k is updated with a small value ΔK as $f\left(x_i,\left(k+\Delta K\right)\right)$. From a Taylor series expansion, it leads to

$$f\left(x_i,\left(k+\Delta K\right)\right)\cong f(x_i,k)+J_i\Delta K \tag{7}$$

where J is a Jacobian matrix (the gradient of $f(x_i,k)$) with respect to k). To minimize the quantity $\left[y_i-f\left(x_i,\left(k+\Delta K\right)\right)\right]^2=\left[y_i-\left(f\left(x_i\right)+J_i\Delta K\right)\right]^2$ the derivative of this function with respect to ΔK is done and it is set the result to zero. This leads to

$$\left(\boldsymbol{J}^T\boldsymbol{J}\right)\Delta K=\boldsymbol{J}^T\boldsymbol{\varepsilon} \tag{8}$$

where bold letters indicate vector quantity. The matrix $\boldsymbol{J}^T\boldsymbol{J}$ in the left hand side of Eq. (8) is Hessian matrix. In Levernberg Marquardt method, Hessian matrix is modified by adding damping term. Eq. (8) for Levernberg Marquardt method is written as

$$\left(\boldsymbol{J}^T\boldsymbol{J}+\mu\boldsymbol{I}\right)\Delta K=\boldsymbol{J}^T\boldsymbol{\varepsilon} \tag{9}$$

where μ is damping and $\boldsymbol{I}$ is the identity matrix. When the damping term is increased, this method, therefore, approaches the steepest descent algorithm. However, if the damping term is decreased to zero, this algorithm is Gauss-Newton method. In Eq. (9), the term ΔK is found and it is used to adjust the value of k .

3. RESULTS AND DISCUSSION

For a certain pressure, the calculation is done at different temperatures. The modified parameters a shown in Eqs. (4) and (5) are found. The coefficients k_1 to k_4 are computed based on the LM method. The coefficients obtained from fitting data are shown in Table 2. In fact, the calculation is conducted from 75 bar to 147.6 bar (P_r = 1.017 to 2.001) with incremental of 5 bar. In Table 2, the results are expressed for some values to give the idea how the coefficients change as pressure changes. From the table, it can be observed that different pressure causes different value of coefficients. For k_1 , it increases as pressure increases. At pressure of 130 bar (P_r =1.762), k_1 shows nearly the same value as the constant in PR equation of state. However, the values of k_2 in considered pressure range are lower when compared with the same constant in PR equation of state. For the constants in exponential form, k_3 is marginally different from k_1 , while k_4 is slightly lower than k_2 at the same pressure.

Table 2 Fitted coefficients for Eqs (4) and (5)

Pressure (bar)	k_1	k_2	k_3	k_4
PR	0.45724	0.71124	-	-
75	0.43780	0.65880	0.43739	0.63843
80	0.43999	0.66180	0.43964	0.64204
90	0.44432	0.66766	0.44410	0.64909
100	0.44824	0.67245	0.44812	0.65484
110	0.45170	0.67612	0.45163	0.65918
120	0.45469	0.67871	0.45465	0.66218
130	0.45722	0.68034	0.45718	0.66398
140	0.45932	0.68111	0.45926	0.66472
147.6	0.46065	0.68119	0.46056	0.66466

To evaluate the accuracy of the modified function, Table 3 shows absolute values of average percent relative error, called AARE%, which is defined as:

$$AARE\%=\left(\frac{1}{N}\sum_{i=1}^N\left|\frac{\rho_i^{SW}-\rho_i^{cal}}{\rho_i^{SW}}\right|\right)\times 100 \tag{10}$$

It is found that the calculation of density using a_{MPR1} gives more accurate value than that using PR equation of state in some pressure rages. At 75 bar, AARE% for using a_{MPR1} in density calculation is 0.581, while that for using PR equation of state is 0.703. At pressure of 130 bar or higher, the modified PR equation of state using a_{MPR1} gives the better calculation result when compared with using original PR equation of state. It is worth to emphasis here before further discussion that AARE% indicates the *average* value of relative errors of considered data. As the calculation data are carefully observed, At near critical temperature, the PR equation of state can provide more accurate value of density than that computed by the modified PR equation of state using a_{MPR1}. At a few degrees above critical temperature, using a_{MPR1} provides density value much diverse from SW equation of state when compared with PR equation of state. Even through there are a few data points of inaccurate predicted density, these high errors of density calculation clearly raise the value of AARE%. For the case of a_{MPR2} , the values of AARE% show slightly lower than that of a_{MPR1} and the same behavior, as discussed above for a_{MPR1} cases, is also observed in a_{MPR2} cases. Moreover, it is worth to discuss here that the least square cure fitting with LM algorithm is a numerical technique which provides the *best result*. It means that the total error from fitting all data should be lower as much as possible. However, this technique may not provide completely fitted results in all rages, especially in complicated problem, and the accuracy also depends on the form of equation used to mimic the behavior. Therefore, the coefficients given by this method can provide good predicted results in some rages, but, in other rages, it may give moderately good predicted results.

The value of coefficient of determination $\left(R^2\right)$, defined in Eq. (11), is found that it varies between 0.718 to 0.997 for both a_{MPR1} and a_{MPR2} cases. In some pressure ranges, R^2's are not satisfy in the researcher's opinion. The accuracy improvements for a_{MPR1} and a_{MPR2} are further discussed in the next section. Then, the concepts of modified parameter a are finally summarized.

$$R^2 = 1 - \frac{\sum_{i=1}^{N}\left(\rho_i^{SW} - \rho_i^{cal}\right)^2}{\sum_{i=1}^{N}\left(\rho^{mean} - \rho_i^{cal}\right)^2} \tag{11}$$

Table 3 Comparison of predicted density using a_{MPR1}, a_{MPR2}, and a in PR equation of state

Pressure (bar)	AARE%		
	a_{MPR1}	a_{MPR2}	a
75	0.581	0.576	0.703
80	0.965	0.959	0.754
90	1.226	1.214	0.805
100	1.172	1.155	0.821
110	1.071	1.049	0.834
120	0.965	0.940	0.866
130	0.862	0.835	0.932
140	0.764	0.737	1.075
147.6	0.693	0.668	1.198

3.1 Improvement of accuracy

As discussed in the previous section, the modifications of parameter a following Eqs.(4) and (5) provide the unsatisfactory density prediction near critical temperature, while PR equation of state can give better predicted result in the same range of temperature. From predicted density delivering by both modified PR equations of state as well as PR equation of state, the absolute relative error values of both modified PR equations of state reduce and approach to that of PR equation of state when the reduced temperature is about 1.08 or at temperature of about 328.5 K. Thus, the concept of improving accuracy is that the density prediction in the temperature range from reduced temperature of 1.00 to 1.08, should uses PR equation of state. If the density prediction for reduced temperature higher than 1.08 is desired, the coefficients k_1 and k_2 for Eq. (4) or the coefficients k_3 and k_4 for Eq. (5) should be used. To show the error from using the four fitted coefficients for density prediction in new temperature range (328.5K-700K), the values of AARE% for density prediction associated with temperature of 328.5K-700K are presented in Table 4.

From Table 4, it is clearly seen that the values of AARE% for modified PR equations are improved from that shown in Table 3. The data at P=120 bar is selected, as an example, to show the deviation of the results from the modified equation compared to those from SW equation. Due to large amount of data, only some deviations of the results are presented and those can be found in Appendix. However, the values of AARE% for using a_{MPR1} and a_{MPR2} at 120 and 130 bar are higher than that for using a. At 110 bar, a_{MPR1} is detected that it has moderately higher AARE% than PR equation of state case. Except these pressures, the modified equations yield obviously lower AARE% than PR equation of state case. Thus, the summary of the concept, obtained from this section, is that to predict the density of CO$_2$, the use of coefficients k_1 and k_2 for Eq. (4) or k_3 and k_4 for Eq. (5), shown in Table 2, are recommended for density prediction in the temperature range of 328.5K-700K. For lower temperature, PR equation of state can be employed for the purpose.

Table 4 Comparison of density prediction in temperature range of 328.5K-700K

Pressure (bar)	AARE%		
	a_{MPR1}	a_{MPR2}	a
75	0.201	0.216	0.594
80	0.230	0.245	0.591
90	0.302	0.314	0.553
100	0.397	0.407	0.458
110	*0.508*	0.414	0.414
120	*0.596*	*0.601*	0.474
130	*0.630*	*0.632*	0.599
140	0.622	0.621	0.805
147.6	0.599	0.597	0.974

For developing computer code, it is more convenient to generate the coefficients, used in calculation, as equation. The coefficients k_1 to k_4, expressed in Table 2, which are weakly dependent on pressure, are formed as functions of reduced pressure. Heidaryan and Jarrahian [11] have modified Redlich Kwong Equation of state. They fitted β function as a function of reduced temperature and reduced pressure. Thus, the coefficients k_1 to k_4 in this study are found as function of reduced pressure. Fig. 1 illustrates the variation of k_1 and k_3 at different reduced pressures, while k_2 and k_4 are plotted versus reduced pressure in Fig. 2. In Fig.1, it also presents an equation, which is obtained by fitting data of k_1 and k_3 and cross symbol plotted from the fitting equation, kf_i (i=1, 3). The coefficient of determination in this figure, which equals to 1 and 0.999 for kf_1 and kf_3, respectively, can be implied that the fitting equation completely relates to the data of k_1 and k_3. For the coefficients k_2 and k_4, these are fitted and the equations are expressed in the figure with the coefficient of determination of 1.0 for both equations.

3.2 Validation

To verify that the modified PR equation of state can be used, even outside fitting range, the modified PR equations of state is employed to find the density at higher pressure and temperature, which are above fitting range. The calculation densities are compared with the densities obtained from SW equation of state with the same pressure and temperature. The temperature range of 700 to 1200 K and the pressure range of 160 to 250 bar are selected to investigate the density calculation. In each pressure, the temperature is varied from 700 to 1200 K. The density is found and the AARE% and R^2 are figured.

The computation results show that the modified PR equations of state provides the CO_2 densities at defined pressure and temperature ranges with AARE% of 0.098-0.424 and R^2 of 0.9994-1.0. The highest AARE% takes place at 250 bar for both cases of modified equations of state. Even though it is highest AARE% in this test, it is less than haft percent. Moreover, for all test cases, AARE% and R^2 from the modified PR equations of state have better values than that from PR equation of state. To illustrate the estimation of density by modified PR Equation of state and PR Equation of state, Fig. 3(a) presents the comparison of density calculated by both modified equations of state and that by SW equation of state at 200 bar. Comparing with the density calculated from PR equation of state (Fig. 3(b)), it can be observed that at higher density (lower temperature), PR equation of state estimates slightly lower value of density. The modified PR equations of state can give more accurate density, when it is compared with the original PR Equation of state. This may be because the outside fitting rage is not highly nonlinear relation of $P\rho T$ when compared with that near critical region. Moreover, the fitted data at the upper bound of fitting rage can also represent the behavior of outside fitting rage.

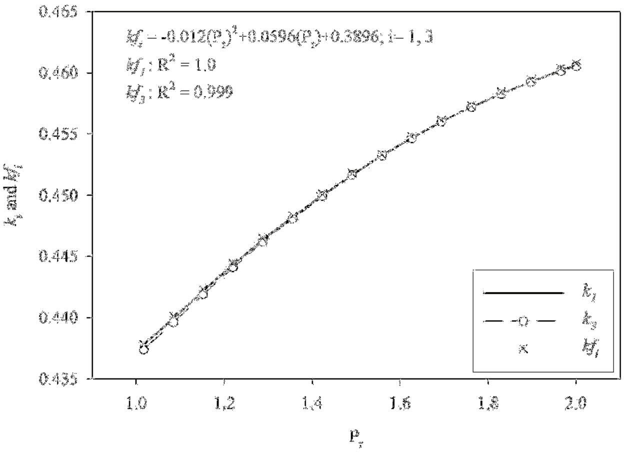

Figure 1 The variations of k_1 and k_3 at different reduced pressures

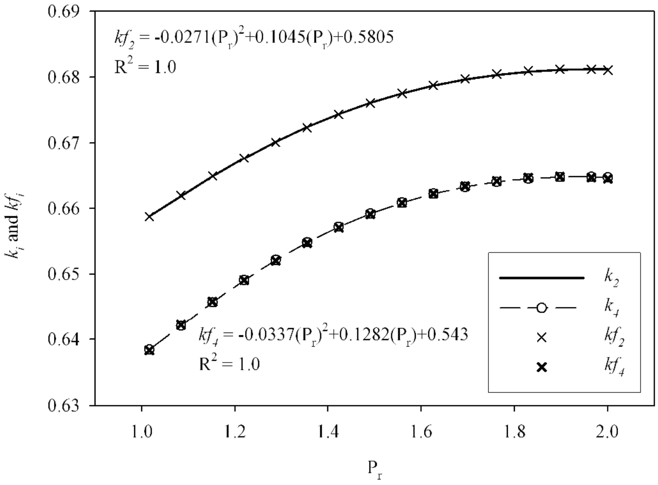

Figure 2 The variations of k_2 and k_4 at different reduced pressures

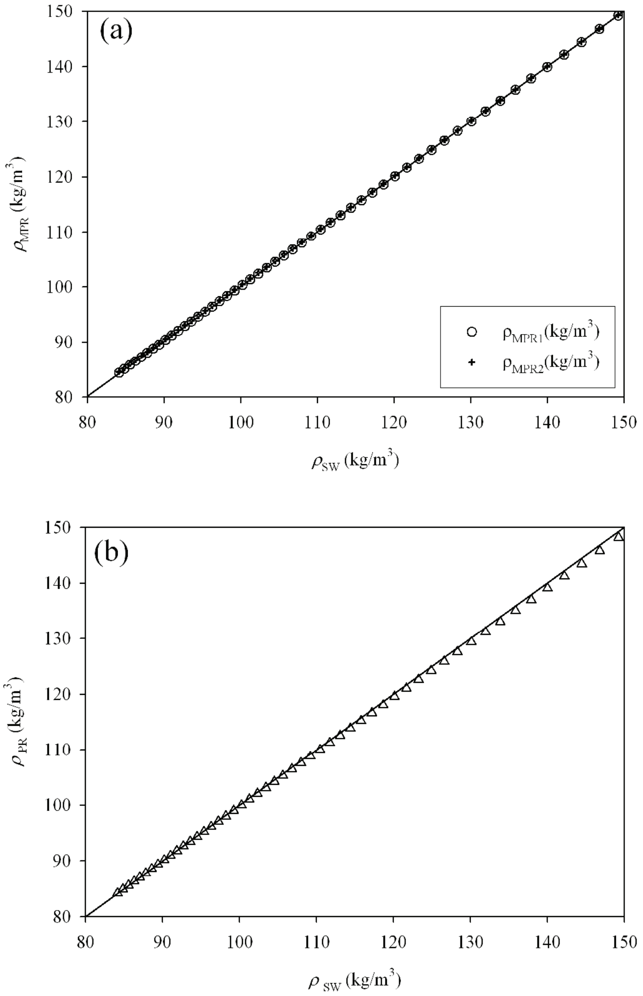

Figure 3 the comparison of calculated density at 200 bar(a) between modified PR equations of state and SW equation of state (b) between PR equation of state and SW equation of state

To compare the predicted density with another equation, SRK equation of state is used to calculate the density. As expressed in Table 5, AARE% for SRK equation is shown and compared with that from modified models. It is found that AARE% from the modified models are generally lower than that from SRK equation.

Table 5 Comparison of predicted density using modified models and SRK equation of state

Pressure (bar)	AARE%		
	a_{MPR1}	a_{MPR2}	a_{SRK}
160	0.110	0.144	1.643
180	0.101	0.163	1.811
200	0.101	0.211	1.970
220	0.180	0.328	2.120
240	0.332	0.403	2.260
250	0.424	0.423	2.327

4. CONCLUSIONS

In this work, two forms of new parameter a in Peng-Robinson (PR) equation of state were proposed for supercritical CO$_2$. The first one used the same form as PR equation, but the coefficients were modified. The second modification is using exponential form for parameter a. The coefficients in both forms were fitted with $P\rho T$ data obtained from Spang-Wagner equation of state by using Levernberg-Marquardt method. The fitted coefficients were weakly dependent on pressure. The comparison result showed that, the density predicted by using modified PR equations of state can usually provide more accurate result than the original PR equation of state in supercritical region. The modified equations were recommended to use for the temperature of 328.5K or higher and the pressure of 75 bar or higher with highest AARE% of 0.630 for the first modification and 0.632 for the second modification. Moreover, the modified equations can also predict the density at outside fitting ranges of pressure and temperature with high accuracy. Use of PR equation is suggested because it is simple. However, the modified models are recommended to use in some range for getting better results.

5. APPENDIX

A1 Span and Wagner equation of state

In Span and Wagner equation on state, the properties of CO$_2$ are expressed in terms of the dimensionless Helmholtz free energy, ϕ, and its derivative. The dimensionless Helmholtz free energy is split into a part depending on the ideal behavior, ϕ^o, and a part taking into account the residual fluid behavior, ϕ^r, as shown in Eq. (A1).

$$\phi(\delta,\tau)=\phi^o(\delta,\tau)+\phi^r(\delta,\tau) \qquad \text{(A1)}$$

where τ is the inverse reduced temperature and δ is the reduced density. Both are defined as:

$$\tau=\frac{T_c}{T}, \quad \delta=\frac{\rho}{\rho_c} \qquad \text{(A2)}$$

where T_c and ρ_c are the critical temperature and the critical density, respectively. The formulations, used to compute the ideal gas part and the residue part of Helmholtz free energy, are:

$$\begin{aligned}\phi^o(\delta,\tau) &= \ln(\delta)+a_1^o+a_2^o\tau+a_3^o\ln(\tau)+ \\ &\sum_{i=4}^{8} a_i^o \ln\left[1-\exp\left(-\tau\theta_i^o\right)\right]\end{aligned} \qquad \text{(A3)}$$

$$\begin{aligned}\phi^r &= \sum_{i=1}^{7} n_i\delta^{d_i}\tau^{t_i} + \sum_{i=8}^{34} n_i\delta^{d_i}\tau^{t_i}\exp(-\delta^{c_i})+ \\ &\sum_{i=35}^{39} n_i\delta^{d_i}\tau^{t_i}\exp\left[-\alpha_i(\delta-\varepsilon_i)^2-\beta_i(\tau-\gamma_i)^2\right]+ \\ &\sum_{i=40}^{42} n_i\Delta^{b_i}\delta\exp\left[-C_i(\delta-1)^2-D_i(\tau-1)^2\right]\end{aligned} \qquad \text{(A4)}$$

where

$$\Delta=\left\{(1-\tau)+A_i\left[(\delta-1)^2\right]^{1/(2\beta)_i}\right\}^2+B_i\left[\left((\delta-1)^2\right)\right]^{a_i} \quad .$$

The coefficients $a_1^o-a_8^o$ and n_i and the parameters θ_i^o, d_i, t_i, c_i, α_i, β_i, γ_i, ε_i, A_i, B_i, C_i, and D_i, totally 201 terms, can be found in Ref. [10].

To find the pressure, the following equation is used:

$$\frac{P(\delta,\tau)}{\rho RT}=1+\delta\frac{\partial\phi^r}{\partial\delta} \qquad \text{(A6)}$$

where R is the universal gas constant. For other properties, such as enthalpy and entropy, their deatils can be found in Ref. [10].

A2 Comparison of predicted density from modified models and SW equation

Table A1 shows the deviation of the results from modified equations and from PR equation compared with those from the SW equation, associated with the data in Table 4. Only the case of P=120 bar is shown as an example. Due to large number of data, only some results are presented in this table. The deviation $\Delta MPR1$ is $\left|\rho^{SW}-\rho^{MPR1}\right|$ and this meaning is also applied for $\Delta MPR2$ and ΔMPR.

Table A1 The deviation of the results from modified equations and from PR equation compared with those from the SW equation

T (K)	$\Delta MPR1$ (kg/m^3)	$\Delta MPR2$ (kg/m^3)	ΔMPR (kg/m^3)	T (K)	$\Delta MPR1$ (kg/m^3)	$\Delta MPR2$ (kg/m^3)	ΔMPR (kg/m^3)
306	49.001	49.004	39.878	478	0.548	0.209	0.046
308	52.845	52.861	43.717	480	0.529	0.194	0.031
310	56.466	56.503	47.349	482	0.510	0.177	0.016
312	59.716	59.785	50.641	484	0.492	0.162	0.001
314	62.384	62.496	53.395	486	0.474	0.147	0.013
316	64.191	64.357	55.353	488	0.456	0.132	0.027
318	64.809	65.039	56.216	490	0.439	0.118	0.040
320	63.896	64.197	55.671	492	0.423	0.104	0.053

T (K)	$\Delta MPR1$ (kg/m^3)	$\Delta MPR2$ (kg/m^3)	ΔMPR (kg/m^3)	T (K)	$\Delta MPR1$ (kg/m^3)	$\Delta MPR2$ (kg/m^3)	ΔMPR (kg/m^3)
322	61.125	61.501	53.419	494	0.406	0.090	0.066
324	56.315	56.764	49.285	496	0.391	0.078	0.078
326	49.719	50.229	43.495	498	0.376	0.065	0.090
328	42.086	42.643	36.739	500	0.360	0.052	0.102
330	34.340	34.928	29.868	502	0.345	0.040	0.113
332	27.254	27.858	23.593	504	0.331	0.029	0.124
334	21.254	21.865	18.307	506	0.316	0.017	0.135
336	16.413	17.023	14.072	508	0.303	0.007	0.145
338	12.599	13.204	10.760	510	0.289	0.004	0.156
340	9.617	10.215	8.191	512	0.276	0.015	0.166
342	7.288	7.877	6.199	514	0.263	0.025	0.175
344	5.461	6.042	4.650	516	0.251	0.034	0.184
346	4.022	4.595	3.438	518	0.238	0.045	0.193
348	2.881	3.446	2.485	520	0.227	0.053	0.201
350	1.970	2.527	1.730	522	0.215	0.063	0.210
352	1.241	1.791	1.130	524	0.203	0.071	0.218
354	0.653	1.197	0.652	526	0.192	0.080	0.226
356	0.177	0.714	0.268	528	0.181	0.088	0.234
358	0.208	0.323	0.040	530	0.171	0.095	0.241
360	0.521	0.005	0.286	532	0.160	0.104	0.249
362	0.776	0.255	0.485	534	0.150	0.111	0.256
364	0.981	0.465	0.642	536	0.141	0.118	0.262
366	1.148	0.637	0.768	538	0.132	0.124	0.268
368	1.283	0.776	0.867	540	0.122	0.131	0.275
370	1.391	0.888	0.944	542	0.113	0.138	0.281
372	1.476	0.977	1.003	544	0.104	0.144	0.286
374	1.543	1.047	1.047	546	0.095	0.150	0.292
376	1.593	1.102	1.078	548	0.087	0.156	0.297
378	1.630	1.142	1.098	550	0.079	0.161	0.302
380	1.657	1.173	1.111	552	0.071	0.167	0.308
382	1.674	1.193	1.115	554	0.063	0.173	0.313
384	1.682	1.205	1.114	556	0.055	0.178	0.318
386	1.684	1.210	1.107	558	0.048	0.182	0.322
388	1.681	1.210	1.096	560	0.041	0.187	0.326
390	1.673	1.204	1.081	562	0.034	0.191	0.330
392	1.660	1.195	1.064	564	0.027	0.196	0.335
394	1.644	1.182	1.044	566	0.021	0.199	0.338
396	1.625	1.166	1.021	568	0.015	0.203	0.342
398	1.604	1.148	0.997	570	0.008	0.208	0.346
400	1.581	1.128	0.972	572	0.002	0.211	0.349
402	1.556	1.106	0.947	574	0.004	0.215	0.352
404	1.529	1.082	0.919	576	0.009	0.218	0.355
406	1.502	1.058	0.892	578	0.015	0.222	0.359
408	1.473	1.032	0.863	580	0.020	0.224	0.361
410	1.444	1.006	0.835	582	0.025	0.227	0.364
412	1.414	0.979	0.806	584	0.031	0.231	0.367
414	1.384	0.952	0.777	586	0.035	0.232	0.369
416	1.354	0.925	0.749	588	0.041	0.235	0.372
418	1.323	0.897	0.720	590	0.045	0.238	0.374
420	1.292	0.869	0.691	592	0.050	0.240	0.376
422	1.262	0.842	0.664	594	0.055	0.242	0.378
424	1.231	0.814	0.635	596	0.058	0.244	0.380
426	1.201	0.786	0.607	598	0.063	0.246	0.382
428	1.171	0.759	0.580	600	0.067	0.248	0.384
430	1.142	0.733	0.554	602	0.071	0.250	0.385
432	1.112	0.706	0.527	604	0.075	0.251	0.386
434	1.083	0.680	0.501	606	0.078	0.253	0.388
436	1.054	0.654	0.475	608	0.082	0.254	0.389
438	1.025	0.628	0.450	610	0.085	0.255	0.390
440	0.996	0.602	0.425	612	0.089	0.257	0.391
442	0.969	0.578	0.401	614	0.091	0.257	0.392
444	0.942	0.554	0.377	616	0.095	0.259	0.394
446	0.916	0.530	0.354	618	0.098	0.260	0.394
448	0.889	0.507	0.331	620	0.101	0.260	0.395
450	0.863	0.483	0.309	622	0.103	0.261	0.395
452	0.837	0.461	0.287	624	0.106	0.262	0.396
454	0.812	0.439	0.266	626	0.109	0.263	0.397
456	0.788	0.417	0.245	628	0.111	0.263	0.397
458	0.764	0.396	0.225	630	0.114	0.263	0.397
460	0.740	0.375	0.205	632	0.116	0.264	0.398
462	0.717	0.355	0.185	634	0.118	0.264	0.398
464	0.695	0.336	0.167	636	0.121	0.265	0.398
466	0.672	0.316	0.148	638	0.122	0.264	0.398
468	0.650	0.297	0.130	640	0.125	0.264	0.398
470	0.629	0.279	0.112	642	0.127	0.265	0.398
472	0.608	0.261	0.095	644	0.129	0.265	0.398
474	0.588	0.244	0.079	646	0.131	0.265	0.399
476	0.567	0.226	0.062	648	0.132	0.264	0.398
650	0.134	0.264	0.398	676	0.149	0.256	0.390
652	0.136	0.264	0.398	678	0.151	0.256	0.390
654	0.137	0.264	0.397	680	0.151	0.254	0.388
656	0.138	0.263	0.397	682	0.152	0.254	0.388
658	0.140	0.263	0.396	684	0.153	0.253	0.386
660	0.141	0.262	0.395	686	0.153	0.252	0.386
662	0.143	0.262	0.396	688	0.154	0.251	0.385
664	0.144	0.261	0.395	690	0.154	0.250	0.383
666	0.145	0.260	0.394	692	0.154	0.248	0.382
668	0.146	0.260	0.393	694	0.155	0.247	0.381
670	0.147	0.259	0.393	696	0.156	0.246	0.380
672	0.148	0.259	0.392	698	0.156	0.245	0.379
674	0.148	0.257	0.391	700	0.157	0.244	0.378

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