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Predicting Natural Gas Dew Points from 15 Equations of State

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This paper is a comparative study of 15 cubic equations of state (EoSs) for predicting natural gas dew points. Two-, three-, and four-parameter EoSs are used to predict the natural gas dew points. Natural gases contain a large amount of supercritical methane; therefore, the fugacity of methane, as a test of suitability of the EoSs, is predicted and compared to the recommended values in International Union of Pure and Applied Chemistry (IUPAC) tables. The vapor pressures of components that are normally present in natural gas mixtures, as another test of the suitability of the EoSs, are also predicted and compared with experimental data. The dew points of several natural gas mixtures then are predicted, using the EoSs, and compared to experimental values. Results reveal that the dew points of lean synthetic natural gases are predicted best by the Redlich–Kwong family of EoSs, whereas rich natural gases dew points are described significantly better by the Patel–Teja family of EoSs.

1. Introduction

Predicting the natural gas dew point is of primary importance in reservoir engineering, where liquid drop-out diminishes production from gas condensate wells. It is also equally important to consider it in the design of gas transportation pipelines and custody transfer.

Equations of state (EoSs) are often used to predict natural gas dew points. Although EoSs are plentiful, the Redlich and Kwong¹ (RK) EoS, a modified version of it by Soave² (RKS), and the Peng and Robinson³ (PR) EoS are usually prescribed for calculating natural gas dew points.⁴ The Patel and Teja⁵ (PT) EoS and a modified version of it by Valderrama⁶ (PTV) are also considered valuable in hydrocarbon processing.^{7,8} However, many EoSs, including some variants of the popular PR and RKS EoSs, have been developed, and this trend seems to be continuing. The question is this: Are the RKS and PR EoSs still the best for hydrocarbon processing? To answer this question, the newly devel-

oped EoSs should be taken into consideration. Clearly, evaluation of all developed EoSs may not be practical,⁹ however, it is likely that, based on some criteria, one can select the appropriate EoSs and study them to calculate natural gas dew points. The subject of this study is to evaluate and recommend suitable EoSs.

Natural gases consist of many components. Nitrogen (N₂), carbon dioxide (CO₂), and hydrogen sulfide (H₂S) are usually the non-hydrocarbon components. Methane (CH₄), ethane (C₂H₆), and other hydrocarbons (up to C₄₀ or even C₅₀) might be present in a natural gas mixture. Nitrogen and methane are always supercritical under reservoir conditions, whereas the heavy hydrocarbons, which cause condensation, are present under subcritical conditions. The temperature and pressure of a hyperbaric reservoir¹⁰ could be as high as 450 K and 100 MPa, respectively. In fact, any EoS capable of predicting natural gas dew points should be able to characterize these complex mixtures under reservoir conditions accurately. The following strategy is then outlined: Only cubic EoSs are evaluated. These EoSs are empirical or semiempirical, yet simple and robust, and their application to natural gas mixtures has been proven useful. Because the RKS and PR EoSs have the best pressure–volume–temperature (PVT) relationships among the other two-parameter EoSs, several variants of them will be studied. Three-parameter EoSs and the

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Table 1. PVT Relations for the Equations of State (EoSs) Used in This Study

equation of state, EoS	PVT relation ^a
RKS, RKT, RKSS	$P = \frac{RT}{v-b} - \frac{a_c \alpha(T_r)}{v(v+b)}$
PR76, PR78, PRD, PRT, PRF, PRG	$P = \frac{RT}{v-b} - \frac{a_c \alpha(T_r)}{v(v+b) + b(v-b)}$
PT, PTV	$P = \frac{RT}{v-b} - \frac{a_c \alpha(T_r)}{v(v+b) + c(v-b)}$
SW	$P = \frac{RT}{v-b} - \frac{a_c \alpha(T_r)}{v^2 + (1+3\omega)bv - 3\omega b^2}$
GD	$P = \frac{RT}{v-b} - \frac{a_c \alpha(T_r)}{v(v+c) + c(v-b)}$
MMM	$P = \frac{RT}{v} \left(\frac{v+\epsilon b}{v-b} \right) - \frac{a_c \alpha(T_r) \sqrt{T}}{v(v+N\epsilon b)}$
TBS	$P = \frac{RT}{v-b} - \frac{a_c \alpha(T_r)}{v^2 + (b+c)v - (bc+d^2)}$

^a Calculated using the following parameters: R is the gas constant; T is the temperature; T_r is the reduced temperature; P is the pressure; v is the molar volume; c , d , N , and ϵ are the EoSs parameters; ω is the acentric factor; b is the molecular co-volume; a_c is the attractive parameter at the critical temperature; and α is the attractive parameter temperature dependence (this latter parameter is also called the α -function).

Treble–Bishnoi–Salim¹¹ (TBS) EoS, as the only four-parameter EoS, will also be used to predict natural gas dew points. van der Waals mixing rules with a geometric mean average combining rule for the attractive parameter are used to extend the EoSs to multicomponent mixtures. In most cases, the van der Waals mixing rules adequately describe natural gas mixtures. However, because these mixing rules are not perfect, binary interaction parameters (BIPs) are usually incorporated to correct the interaction between binary molecules; these corrections are represented by the parameter k_{ij} in the EoSs. The impact of binary interaction parameters will also be discussed in this study.

First, the EoSs are used to predict the supercritical behavior of methane and nitrogen, in terms of fugacity, for wide ranges of pressure and temperature. The EoSs then are used to predict the vapor pressure of some light and heavy hydrocarbons at the prevailing temperatures of the reservoirs. The behavior of components in a mixture is clearly different than that in a pure state; however, these comparisons can still give good indications for the suitability of EoSs in predicting natural gas dew points. Afterward, the accuracies of the EoSs are compared to predict the dew points of some synthetic natural gases that were reported recently by a Spanish group.^{12,13} Finally, having specified an exponential decay function for the composition of single carbon number (SCN) groups in C_{7+} fractions, the dew points of rich natural gases are predicted and compared with experimental data. The effect of correlations on determining the critical properties of the SCN group will also be discussed.

2. The Equations of State

The EoSs used in this study are classified as two-, three-, and four-parameter EoSs. The two-parameter EoSs are solely comprised of the RK and PR families.

From the RK family, the modified versions of Soave² (RKS), Twu et al.¹⁴ (RKT), and Souahi et al.¹⁵ (RKSS) are used. From the PR family, the original Peng and Robinson (PR76,³ PR78¹⁶) EoSs are used, as well as modified versions of them by Danesh et al.¹⁷ (PRD), Twu et al.¹⁸ (PRT), Flöter et al.¹⁰ (PRF), and Gasem et al.¹⁹ (PRG). The three-parameter EoSs include the following: Schmidt and Wenzel²⁰ (SW), Guo and Du²¹ (GD), Patel and Teja⁵ (PT), a modified PT EoS by Valderrama⁶ (PTV), and an EoS by Mohsen-Nia, Modarress, and Mansoori²² (MMM). The TBS EoS is the only four-parameter EoS that is used in this study.

The PVT relationships for the previously mentioned EoSs are given in Table 1. In Table 2, the temperature dependences for the attractive terms of the EoSs (which are also called α -functions) are presented. The EoSs are presented briefly in the following sections, however, the details can be found in the given references.

2.1. Two-Parameter Equations of State. 2.1.1. The RKS EoS.²

The RK EoS successfully relates the PVT of

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Table 2. The α -Functions Used in This Study

equation of state, EoS	α -function ^a
RKS, PR76, PR78, PT, PTV, SW, GD, MMM	$\alpha = [1 + m(1 - \sqrt{T_r})]^2$
PRD	$\begin{cases} \alpha_{\text{Subcritical}} = [1 + m(1 - \sqrt{T_r})]^2 \\ \alpha_{\text{Supercritical}} = [1 + 1.21m(1 - \sqrt{T_r})]^2 \end{cases}$
PRF	$\begin{cases} \alpha_{\text{Supercritical methane}} = a_0 + a_1T_r + a_2T_r^2 + a_3T_r^3 \\ \alpha_{\text{Others}} = [1 + m(1 - \sqrt{T_r})]^2 \end{cases}$
PRG	$\alpha = \exp[(A + BT_r)(1 - T_r^{C+D\omega+E\omega^2})]$
RKT, PRT	$\alpha = \alpha^{(0)} + \omega(\alpha^{(1)} - \alpha^{(0)})$
RKSS	$\alpha^{(i)} = T_r^{N(M-1)} \exp[L(1 - T_r^{NM})]$
TBS	$\alpha = \alpha^{(0)} + \omega(\alpha^{(1)} - \alpha^{(0)})$ $\alpha^{(i)} = 1 + [A(1 - T_r) + B(1 - T_r)^2 + C(1 - T_r)^3 + D(1 - T_r)^6]/T_r$ $\alpha = 1 + m(1 - \sqrt{T_r}) + p(\sqrt{0.7} - \sqrt{T_r})(1 - \sqrt{T_r})$

^a Calculated using the following parameters: T_r is the reduced temperature; ω is the acentric factor; and m , a_0 , a_1 , a_2 , a_3 , A , B , C , D , E , N , M , L , and p each are either a constant or a function of the acentric factor.

gases; however, it poorly predicts the vapor pressure and liquid density of pure compounds. Soave² introduced a temperature dependence (α) for the attractive term of the RK EoS, as given in Table 2. This term significantly improves the accuracy of the EoS to predict vapor pressure, although the accuracy of the EoS to predict liquid density remains unchanged.²³ As a consequence of this modification, the RKS can be used successfully in the fluid phase equilibria of hydrocarbon mixtures. However, the α -function causes the RKS to predict anomalous behaviors under extreme conditions.²⁴

2.1.2. The RKT EoS.¹⁴ Soave² correlated the α -function for the RKS EoS by matching the predicted vapor pressure at a reduced temperature of 0.7 to the experimental value. Although it has been proven to be useful, the vapor pressure predicted by the RKS EoS usually gets worse at reduced temperatures of <0.7 . Recognizing this behavior, especially for heavy hydrocarbons, Twu et al.¹⁴ developed an α -function that works well from the triple-point temperature to the critical-point temperature. By predicting the vapor pressure of heavy hydrocarbons well, the RKT EoS could be capable of describing the vapor–liquid equilibria (VLE) of natural gas mixtures that contain heavy hydrocarbons.

2.1.3. The RKSS EoS.¹⁵ Souahi et al.¹⁵ developed a new α -function with the same purpose as Twu et al.;¹⁴ however, their α -function seems to be more accurate in regard to predicting the vapor pressure of pure compounds. However, they did not address the behavior of the α -function under supercritical conditions.

2.1.4. The PR76³ and PR78¹⁶ EoSs. Another successful PVT relation among EoSs is the PR EoS. Compared to RK family of EoSs, the PR family of EoSs generally predicts the liquid density of compounds more accurately;²³ however, the accuracy is not good enough for industrial applications. The PR EoS takes advantage of the Soave-type α -function, hence, showing similar quality with temperature. However, Peng and Robinson³ used a reduced temperature range (0.7–1) to correlate the PR α -function. Also, it was first correlated to pure compounds with an acentric factor of <0.5 (PR76),³ and later, in 1978, they extended it to compounds with a larger acentric factor (PR78).¹⁶

2.1.5. The PRD EoS.¹⁷ The PRD EoS is a revised version of the PR78 EoS. It takes into consideration the

supercritical behavior of components in a hydrocarbon mixture by multiplying the parameter m in the PR α -function by a factor of 1.21. As reported by Danesh et al.,¹⁷ it works well for calculating the dew points of gas condensate mixtures.

2.1.6. The PRF EoS.¹⁰ The PRF is similar to the PRD EoS, i.e., it uses the PR78 EoS α -function for all components except methane under supercritical conditions. Flöter et al.¹⁰ introduced a third-order polynomial α -function for methane under supercritical conditions by matching the fugacity of the PR EoS to the recommended values in IUPAC tables. The PRF EoS is quite impressive for describing the VLE of the binary asymmetric mixtures.

2.1.7. The PRT EoS.¹⁸ The reason for developing the PRT EoS is similar to that for the RKT EoS. The PRT EoS is an improved version of the PR EoS, especially for predicting the vapor pressure of pure compounds at low temperatures.

2.1.8. The PRG EoS.¹⁹ Gasem et al.¹⁹ developed a new α -function in exponential form, having recognized that the Soave-type α -function used by the PR EoS does not decrease monotonically to zero with temperature. The PRG EoS also attempts to improve the predictive capability of the PR EoS for vapor pressure.

2.2. Three-Parameter Equations of State. **2.2.1. The PT⁵ and PTV⁶ EoSs.** The PT and PTV EoSs have the same PVT relationship and α -functions. The difference between them involves the calculation of the EoSs parameters, i.e., a , b , and c . In the PTV EoS, the actual compressibility factor is used, whereas the critical compressibility factor in the PT EoS is a conventional parameter. Consequently, the PTV EoS predicts liquid densities near the critical point more accurately than the PT EoS.

2.2.2. The SW EoS.²⁰ Schmidt and Wenzel²⁰ recognized that the RKS EoS accurately predicts the thermodynamic properties of fluids with an acentric factor of almost zero, whereas the PR EoS with an acentric factor of ~ 0.3 . Knowing that, they developed a new EoS that reduces to the RKS EoS at an acentric factor of 0 and to the PR EoS at an acentric factor of $1/3$. In the SW EoS, the acentric factor is a third parameter. The SW EoS accurately predicts the liquid density and vapor pressure of light and moderate compounds.

2.2.3. The GD EoS.²¹ The GD EoS is a three-parameter EoS that is very similar in form to the PT EoS.⁵ It was developed particularly for hydrocarbon

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mixtures. The α -function for the GD EoS is similar to the Soave-type α -function; however, the parameter m is defined differently for different compounds, based on the acentric factor. As shown by Guo and Du,²¹ the pure and mixture properties are predicted quite accurately using the GD EoS.

2.2.4. THE MMM EoS.²² Mohsen-Nia et al.²² did not use a van der Waals repulsive term for the MMM EoS. Instead, they used a more accurate empirical repulsive term, having considered the molecular simulation data of hard spheres. The MMM EoS is accurate for predicting vapor pressure and liquid density of light and moderate pure compounds. The α -function in the MMM EoS is of the Soave type. They also used a temperature dependence term for the molecular co-volume parameter. As shown by Mohsen-Nia et al.,²² this improves the capability of the EoS to predict liquid density; however, as noted by Salim and Trebble,¹¹ this may cause anomalous behaviors under extreme conditions.

2.3. The Four-Parameter Equation of State: TBS EoS.¹¹ The TBS EoS is a four-parameter EoS. The PVT relationship for the TBS is flexible and can be reduced to other EoSs. Although it is accurate for predicting the thermodynamic properties of pure compounds, its extension to mixtures requires four mixing rules. Considering that mixing rules are generally empirical, the application of four mixing rules may diminish the predictability of the TBS EoS for mixtures.

3. Mixing Rules

The van der Waals mixing rules have proven to be useful in hydrocarbon processing. The van der Waals quadratic mixing rule with a geometric combining rule is used for the attractive parameter of the EoSs:

$$a = \sum_i \sum_j x_i x_j a_{ij} \quad (1)$$

with

$$a_{ij} = \sqrt{a_{ii} a_{jj}} (1 - k_{ij}) \quad (2)$$

where k_{ij} is the BIP. For the second, third, and fourth parameters, the following mixing rule is used:

$$w = \sum_j x_j w_j \quad (3)$$

where $w = b, c, d$, and ω in different EoSs.

4. Heavy Ends

The heavy constituents of a natural gas are usually considered as a group and reported as C_{7+} . The C_{7+} fraction is commonly specified by molecular weight and specific gravity. The C_{7+} fractions may be comprised of many components and different families of hydrocarbons. Although it is possible to analyze the C_{7+} fractions accurately, using new techniques such as gas capillary chromatography, Pedersen et al.²⁵ discovered that the specification of components up to C_{20} may be adequate for an accurate calculation of thermodynamic

properties. Nevertheless, usually, compositions up to C_6 are available with a heavy end that must be characterized.

It is a common practice to split a C_{7+} fraction into several SCN groups and then calculate the critical properties of each group, using available correlations.^{26–29} Katz³⁰ introduced a simple decay exponential function to express the distribution of SCN groups. Pedersen et al.²⁵ also determined that the compositional distribution of SCNs in North Sea petroleum fractions is best-described by an exponential function. Recently, Starling³¹ also suggested an exponential decay function for splitting heavy ends. The decay functions are similar to each other. However, the decay function by Pedersen et al.²⁵ seems to be more general and will be used in this study:

$$Z_n = \exp(A + BM_n) \quad (4)$$

where Z_n is the SCN group mole fraction and M_n is the SCN group molecular weight. The unknowns A and B are determined for a C_{7+} fraction using the following constraints:

$$Z_{C_{7+}} = \sum_{C_7}^{C_N} Z_{C_n} \quad (5)$$

and

$$M_{C_{7+}} = \frac{\sum_{C_7}^{C_N} Z_{C_n} M_{C_n}}{Z_{C_{7+}}} \quad (6)$$

where C_N is the heaviest SCN to be considered in a C_{7+} fraction and C_n is a dummy variable. To determine A and B (and, hence, the SCN distribution), the SCN molecular weight and volume are needed. We used the generalized SCN properties, as reported by Whitson.³²

5. Results and Discussion

The behavior of components in a multicomponent mixture can be significantly different than in a pure state, depending on the difference in size, shape, and polarity of components. However, in the VLE of natural gas mixtures, it is valuable to evaluate (i) the supercritical behavior of methane, which comprises a large portion of natural gases; (ii) the supercritical behavior of nitrogen, which interacts significantly with hydrocarbons; and (iii) the subcritical behavior of hydrocarbons, which cause condensation. This study is limited to temperatures of <450 K and pressures of <100 MPa, because usually reservoir temperatures and pressures fall within these ranges.

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Table 3. Predicting the Fugacity of Methane and Nitrogen from Cubic Equations of State

equation of state, EoS	Methane ^a		Nitrogen ^b	
	%AAD	RMSD (MPa) ^c	%AAD	RMSD (MPa) ^c
RKS	3.77	2.90	2.10	2.86
RKT	6.01	4.68	2.26	2.86
RKSS	7.92	6.91	7.30	11.56
PR76	7.68	6.68	7.96	13.45
PR78	7.51	6.54	7.85	13.31
PRD	22.93	18.95	25.99	34.49
PRF	1.74	1.59	7.85	13.31
PRT	5.19	4.68	5.00	10.30
PT	1.16	0.73	1.58	4.81
PTV	1.46	1.64	2.29	6.10
SW	6.72	5.45	3.37	4.08
GD	2.66	2.27	1.33	3.42
MMM	10.65	8.21	7.18	7.17
TBS	9.84	8.02	12.42	16.39

^a For a temperature range of 220–440 K and pressure range of 1–100 MPa. Data taken from ref 33. Number of points = 96. ^b For a temperature range of 200–450 K and pressure range of 1–100 MPa. Data taken from ref 34. Number of points = 72. ^c Root mean square deviation: $\text{RMSD} = \sqrt{(1/N) \sum_j^N (f_{\text{cald},j} - f_{\text{expl},j})^2}$.

Table 3 presents the average absolute deviation, expressed by

$$\% \text{AAD} = \frac{100}{N} \sum_j^N \frac{|\text{cald}_j - \text{expl}_j|}{\text{expl}_j} \quad (7)$$

where the cald and expl denote the calculated and experimental values for predicting the fugacity of methane and nitrogen for the defined temperature and pressure limits. Under reservoir conditions, methane and nitrogen are always supercritical. The accuracy of an EoS to predict the fugacity of methane leads to the accurate characterization of natural gas mixtures. Indicated in Table 3, the PT, PTV, and GD EoSs are significantly in good agreement with the recommended values in the IUPAC tables.^{33,34} The PRF EoS is also accurate for methane, because the authors matched the fugacity of the EoS to the IUPAC values. Among the two families, the RK family is superior to the PR family and the RKS EoS is remarkably better than the RKT and RKSS EoSs. In the PR family, the PRF and PRT describe the fugacity of methane and nitrogen better than the others. The PRD, MMM, and TBS EoSs are clearly inferior, with respect to the others. Figure 1 depicts the agreement between the PT EoS, as the best EoS in predicting fugacity, and the IUPAC values for the given temperature and pressure ranges.

In Tables 4 and 5, one can see the ability of different EoSs to predict the vapor pressure of potential compounds in natural gases for a temperature range from the triple-point temperature to the critical-point temperature. When the coexistence curve temperature range for a compound was outside of 450 K, the predictions were limited to temperatures of <450 K. The RK family is clearly superior to the PR family. Among the other EoSs, the GD EoS is superior, whereas the

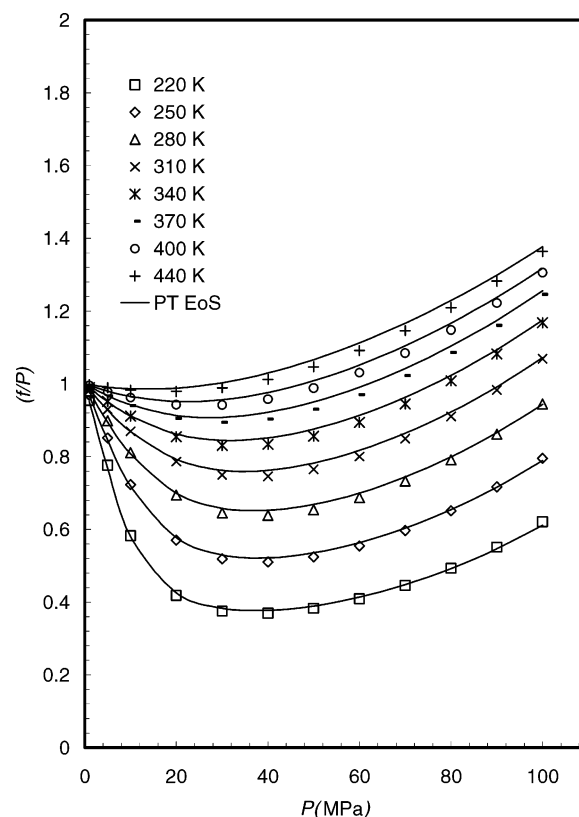


Figure 1. Fugacity coefficients of methane from the PT equation of state (EoS). (Symbols are recommended values from IUPAC.³³)

MMM is inferior. Overall, the RKT and PRT EoSs predict the vapor pressure of pure compounds quite accurately, with the same average absolute deviations. The RKSS and RKS EoSs are ranked third and fourth, respectively. Considering these results and the prediction capability of the EoSs for describing the fugacity of methane, one can conclude that, overall, the RK family EoSs can best predict the VLE of natural gas mixtures and are superior to other EoSs. However, this is not absolutely true, knowing that several other parameters have a role in describing the dew points of natural gases by EoSs.

BIP is one of the important parameters affecting the VLE of mixtures. The BIPs are, at best, empirical, usually temperature-dependent, and dependent on the EoS. The BIPs are determined by correlating (curve-fitting) the VLE of binary mixtures. Having the BIPs of all binaries in a mixture, one can predict the dew points of a mixture and expect a good result. Danesh et al.¹⁷ claimed that, in the VLE of multicomponent mixtures, it is very important to obtain the EoS parameters accurately; however, they never ignore the importance of BIPs in the VLE calculation. In Figure 2, the dew points of a synthetic natural gas mixture are predicted with and without BIPs by the RKS, PR, and PT EoSs and compared to experimental data. The

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Table 4. Predicting the Vapor Pressure of Pure Compounds, in Terms of %AAD, from the Equations of State for the RK and PR Families

component	T_f range	%AAD for Vapor Pressure ^a						
		RKS	RKT	RKSS	PR76	PR78	PRG	PRT
CO ₂	0.71–1	0.44	0.11	0.30	0.52	0.37	0.13	0.11
H ₂ S	0.50–1	2.26	3.69	3.29	3.84	3.76	3.37	3.72
C ₁	0.48–1	2.01	0.26	0.63	0.50	0.52	1.05	0.23
C ₂	0.30–1	3.47	1.83	0.69	3.47	3.35	1.45	1.62
C ₃	0.23–1	3.06	7.25	4.26	9.26	9.92	3.47	6.64
<i>n</i> -C ₄	0.32–1	2.31	2.99	1.71	4.31	4.97	0.85	2.88
<i>i</i> -C ₄	0.28–1	8.92	4.79	8.22	20.09	21.09	10.67	5.34
<i>n</i> -C ₅	0.31–0.96	6.15	2.38	5.04	15.22	16.39	8.67	2.46
<i>i</i> -C ₅	0.25–0.98	2.52	7.81	4.43	11.81	13.19	3.97	7.27
neo-C ₅	0.59–1	0.85	0.37	0.52	0.28	0.30	0.58	0.41
<i>n</i> -C ₆	0.35–0.87	5.36	2.49	3.92	9.60	10.21	5.62	2.58
cyc-C ₆	0.51–0.81	0.60	0.68	0.60	1.38	1.64	1.51	0.80
CH ₃ -cyc-C ₅	0.25–0.84	9.41	4.82	3.17	29.26	31.47	10.03	3.93
C ₆ H ₆	0.50–0.80	0.50	1.36	1.26	2.30	2.64	2.29	1.47
<i>n</i> -C ₇	0.34–0.83	5.46	1.05	3.16	13.69	14.36	7.44	0.98
CH ₃ -cyc-C ₆	0.26–0.79	22.1	4.96	12.99	48.29	51.23	23.11	6.32
<i>n</i> -C ₈	0.38–0.79	2.69	4.67	3.20	4.73	4.77	2.25	4.78
<i>n</i> -C ₉	0.27–0.77	2.78	13.86	8.66	16.25	15.86	2.97	13.41
<i>n</i> -C ₁₀	0.39–0.73	3.84	6.18	4.79	6.87	5.20	3.69	6.41
<i>n</i> -C ₁₂	0.40–0.68	0.67	2.42	1.99	16.08	10.64	5.27	2.72
<i>n</i> -C ₁₄	0.40–0.65	6.32	2.49	6.09	28.64	16.66	9.81	2.16
<i>n</i> -C ₁₆	0.41–0.62	5.89	8.58	2.81	24.71	6.98	3.42	8.48
<i>n</i> -C ₁₈	0.41–0.60	9.35	5.45	13.48	54.48	25.08	16.72	5.46
<i>n</i> -C ₂₀	0.40–0.59	8.54	6.91	13.48	67.76	24.12	16.30	6.98
average		4.81	4.06	4.53	16.39	12.28	6.03	4.05

^a Experimental values taken from ref 35.**Table 5. Predicting the Vapor Pressure of Pure Compounds, in Terms of %AAD, from 6 Cubic Equations of State**

component	T_f range	%AAD for Vapor Pressure					
		PT	PTV	SW	GD	MMM	TBS
CO ₂	0.71–1	0.35	0.23	0.75	1.12	0.15	0.89
H ₂ S	0.50–1	3.38	2.71	3.88	3.57	3.73	2.86
C ₁	0.48–1	1.21	5.48	0.57	0.78	1.06	1.12
C ₂	0.30–1	1.63	3.03	2.39	2.74	1.53	2.52
C ₃	0.23–1	5.65	5.36	5.76	5.04	6.37	2.7
<i>n</i> -C ₄	0.32–1	2.96	5.06	2.27	2.06	2.83	1.24
<i>i</i> -C ₄	0.28–1	16.94	21.54	6.07	6.62	5.57	8.50
<i>n</i> -C ₅	0.31–0.96	13.26	23.52	3.82	4.53	2.73	7.13
<i>i</i> -C ₅	0.25–0.98	8.94	13.79	4.80	3.97	6.87	1.52
neo-C ₅	0.59–1	0.30	0.76	0.29	0.41	0.41	0.58
<i>n</i> -C ₆	0.35–0.87	8.72	11.17	2.90	3.28	3.29	6.09
cyc-C ₆	0.51–0.81	0.99	2.11	0.77	0.69	0.43	0.79
CH ₃ -cyc-C ₅	0.25–0.84	24.43	34.02	1.69	3.35	2.90	10.94
C ₆ H ₆	0.50–0.80	1.84	2.41	1.53	1.43	1.03	1.40
<i>n</i> -C ₇	0.34–0.83	12.45	16.19	2.51	3.09	2.37	9.73
CH ₃ -cyc-C ₆	0.26–0.79	42.18	51.13	11.69	13.98	7.54	25.41
<i>n</i> -C ₈	0.38–0.79	3.43	5.67	3.68	3.48	3.19	4.12
<i>n</i> -C ₉	0.27–0.77	14.55	17.68	7.63	7.66	7.10	3.90
<i>n</i> -C ₁₀	0.39–0.73	4.41	3.32	5.93	4.72	3.45	5.20
<i>n</i> -C ₁₂	0.40–0.68	12.21	7.18	11.29	1.27	13.46	2.95
<i>n</i> -C ₁₄	0.40–0.65	23.70	14.04	26.93	4.90	40.02	3.44
<i>n</i> -C ₁₆	0.41–0.62	19.94	9.17	31.63	7.85	62.74	15.99
<i>n</i> -C ₁₈	0.41–0.60	49.51	13.46	71.08	15.11	142.44	8.82
<i>n</i> -C ₂₀	0.40–0.59	64.55	5.99	109.81	17.97	278.19	16.62
average		14.06	11.46	13.32	4.98	24.97	6.02

compositions of this synthetic natural gas and the others used in this study are given in Table 6. The BIPs have been taken from the literature.^{36,37} As Figure 2 shows, the use of BIPs significantly improves the prediction accuracy of the RKS, PR, and PT EoSs for the dew points of mixture SNG-1. However, in Figure 3, the use of BIPs slightly impairs the predictions for mixture SNG-2. It is also shown that the dew points of mixture SNG-2 have been predicted by the RKA and PTV EoSs. For these predictions, the BIPs of the RKS EoS have been used for the RKA EoS and the BIPs of the PT EoS for the PTV EoS. It may be possible to use the BIPs of

Table 6. Compositions of Lean Synthetic Natural Gases Used in This Study^a

component	Composition (mol %)				
	SNG-1	SNG-2	SNG-3	SNG-4	SNG-5
CO ₂	25.908	0.202	1.7	0.51	0.284
N ₂	1.559	0.313	0.772	6.9	5.651
C ₁	69.114	90.483	84.446	88.1882	83.3482
C ₂	2.62	8.038	8.683	2.72	7.526
C ₃	0.423	0.801	3.297	0.85	2.009
<i>i</i> -C ₄	0.105	0.081	0.293	0.17	0.305
<i>n</i> -C ₄	0.104	0.123	0.589	0.32	0.52
<i>i</i> -C ₅	0.034	0.01	0.084	0.085	0.12
<i>n</i> -C ₅	0.023	0.0079	0.086	0.094	0.144
<i>n</i> -C ₆	0.110	0.0047	0.05	0.119	0.068
<i>n</i> -C ₇		0.0011		0.0258	0.0138
<i>n</i> -C ₈				0.018	0.011

^a Data taken from refs 12 and 13.

one EoS for the other EoS, having compared the predictions with several experimental data.⁷ However, there are other EoSs to be used in this study and the BIPs are not available for them. To have a fair comparison, hereafter, we set a zero value for BIPs for all binaries in EoSs.

Figures 4–6 show the dew points of mixture SNG-3, as predicted by the RK family, PR family, and the other EoSs, respectively, and experimental values are also shown. Clearly, the RK family EoSs are in better agreement with experimental data than those of the PR family and the other EoSs. For the RK family EoSs, regardless of which EoS is used, the predictions are similar. In the PR family, the PRT EoS is somewhat superior. It is also shown that the PRD and PRF EoSs overpredict the upper branch of the dew point curve. Among the three-parameter EoSs, the SW EoS exhibits superiority, whereas the MMM exhibits inferiority. The TBS EoS is comparable to the MMM EoS.

Figures 7–9 illustrate the dew point loci for mixture SNG-4, as predicted by the EoSs and compared to the

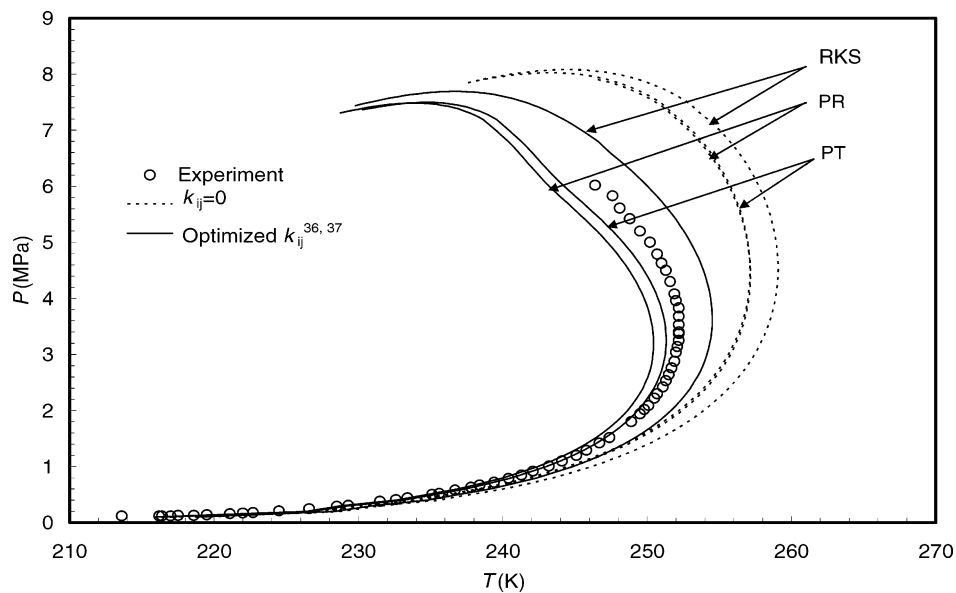


Figure 2. Experimental (from ref 12) and predicted dew points for mixture SNG-1.

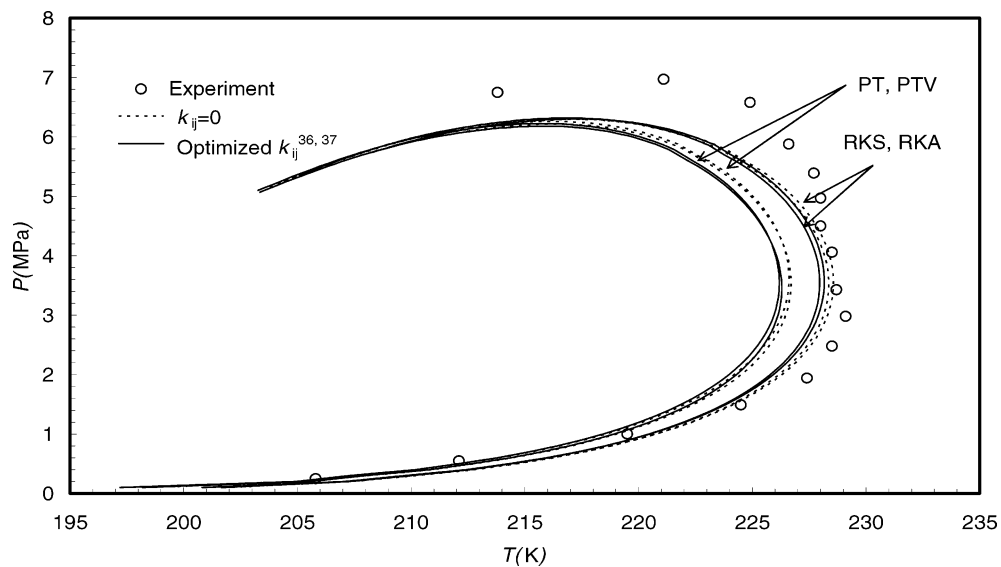


Figure 3. Experimental (from ref 13) and predicted dew points for mixture SNG-2.

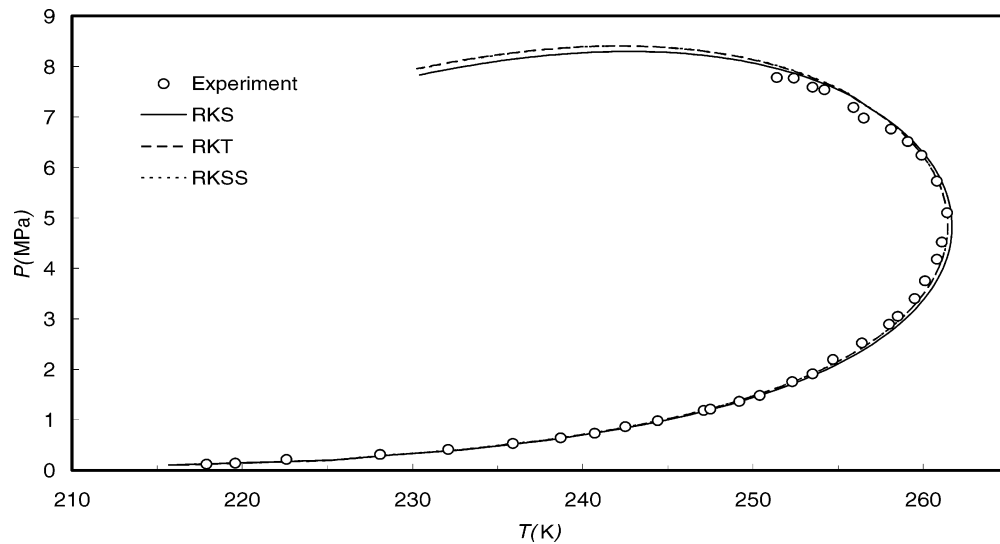


Figure 4. Experimental (from ref 12) and predicted dew points from the RK family for mixture SNG-3.

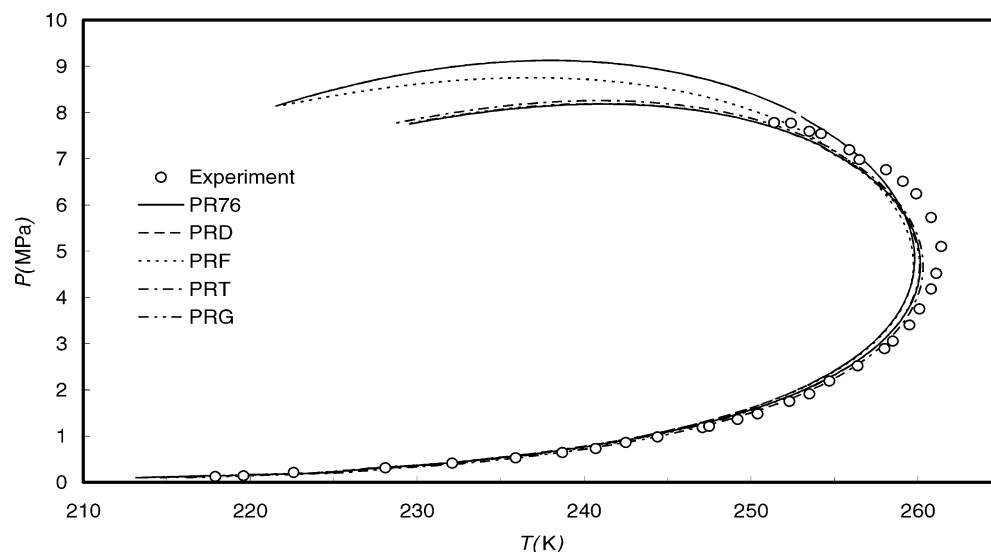


Figure 5. Experimental (from ref 12) and predicted dew points from the PR family for mixture SNG-3.

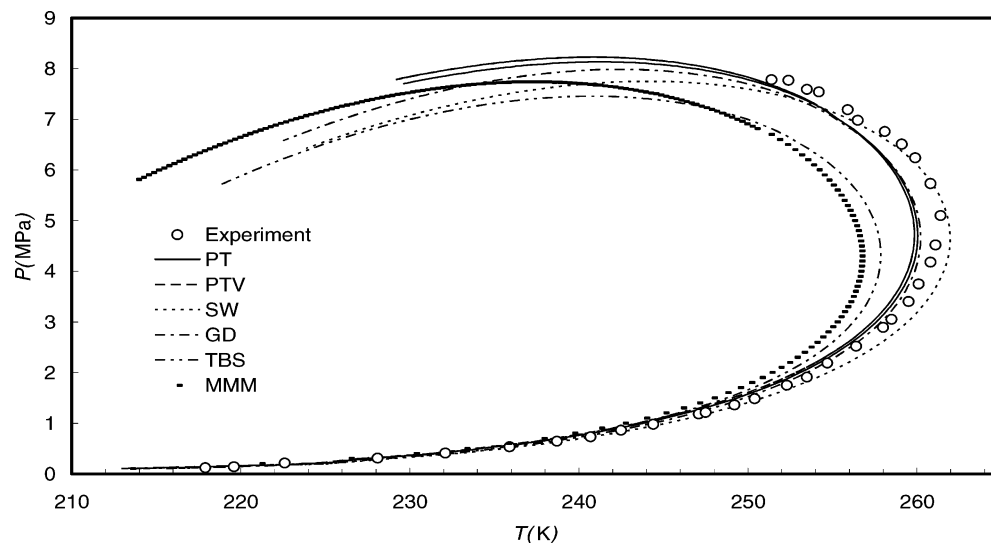


Figure 6. Experimental (from ref 12) and predicted dew points from six EoSs for mixture SNG-3.

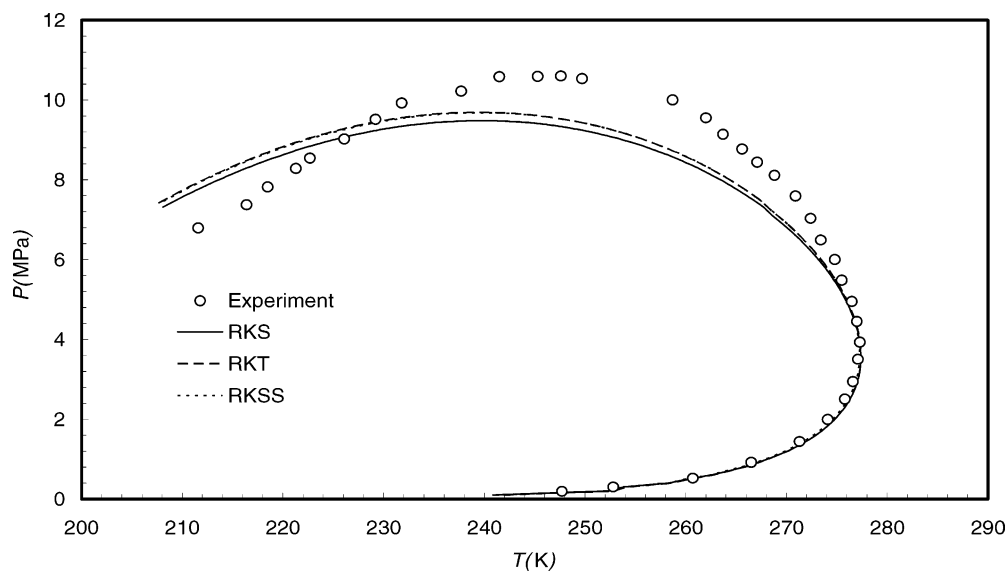


Figure 7. Experimental (from ref 13) and predicted dew points from the RK family for mixture SNG-4.

experimental data. As can be seen, the cricondentherm can only be predicted by the RK family EoSs, and the PRD and SW EoSs; however, except for the PRD EoS,

none of the EoSs are capable of predicting the cricondenbar. The PRD EoS is in good agreement with the experimental data and considerably superior with re-

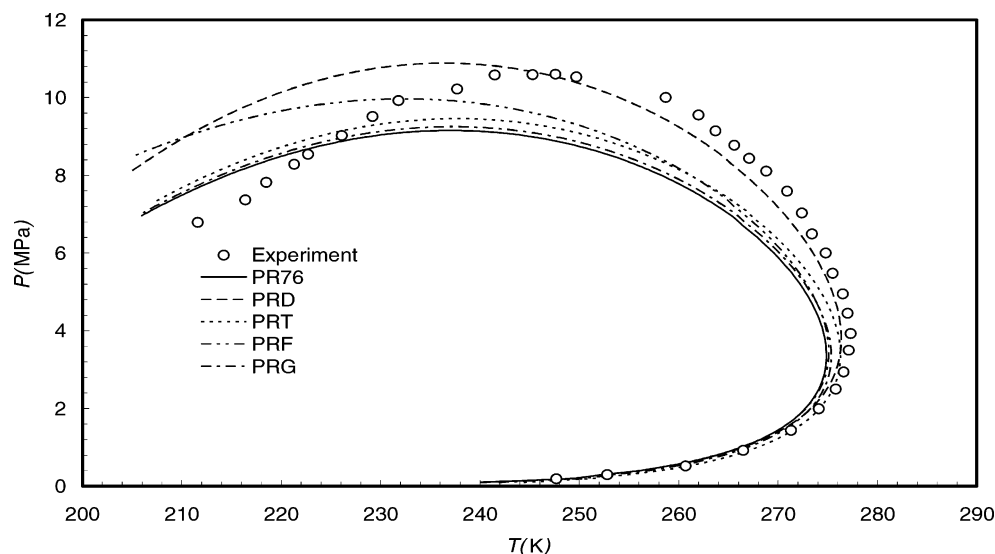


Figure 8. Experimental (from ref 13) and predicted dew points from the PR family for mixture SNG-4.

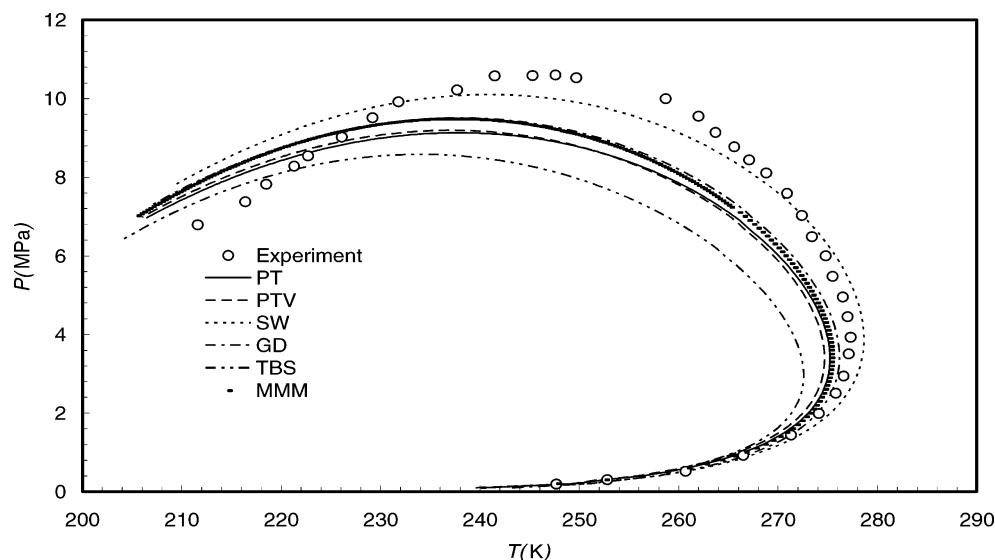


Figure 9. Experimental (from ref 13) and predicted dew points from six EoSs for mixture SNG-4.

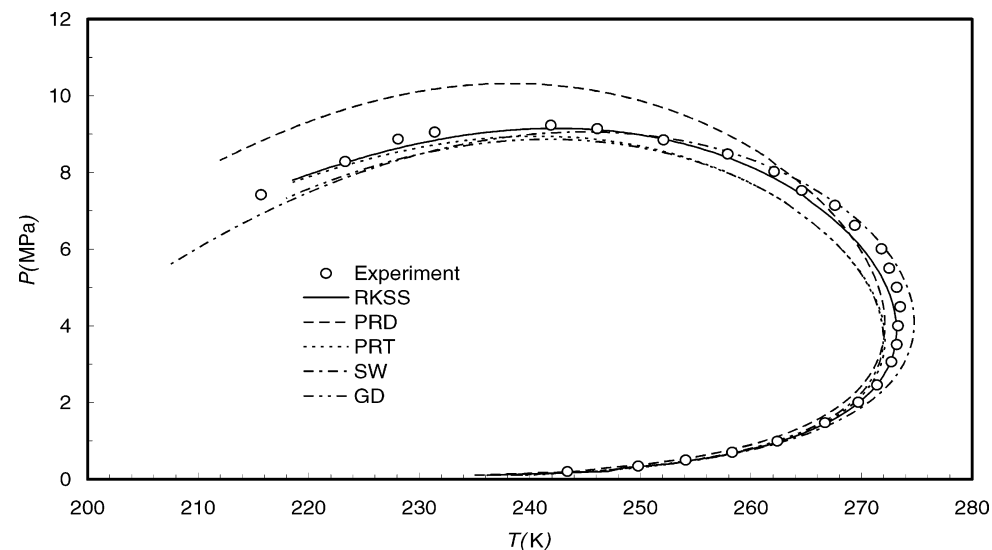


Figure 10. Experimental (from ref 13) and predicted dew points from the best EoS for mixture SNG-5.

spect to the others. However, the RK family EoS, and the PRT, SW, and GD EoSs are also in agreement with the experimental data. The predictions from these EoSs

are also compared with the experimental data for mixture SNG-5 in Figure 10. Except for the PRD EoS, the agreement with the experimental data is quite good

Table 7. Specifications of Rich Natural Gases Used in This Study^a

specification	Value											
	A ³⁸	B ³⁸	C ³⁹	D ³⁸	E ³⁸	F ⁴⁰	G ³⁸	H ⁴¹	I ⁴²	J ⁴³	K ⁴⁴	L ⁴⁵
composition (mol %)												
CO ₂	0.82	3.47		0.45	0.97	0.10	0.18	8.64		0.61	2.44	2.17
N ₂	0.87	0.39		0.38	0.41		0.15	0.71		0.46	0.08	0.34
C ₁	64.04	80.19	82.38	83.00	86.16	95.22	86.57	70.85	91.35	68.64	82.10	70.64
C ₂	10.57	6.28	4.28	3.76	3.55	1.68	3.83	8.53	4.03	13.90	5.78	10.76
C ₃	5.75	2.75	3.51	1.44	1.54	0.91	1.97	4.95	1.53	6.89	2.87	4.94
<i>i</i> -C ₄	1.35	0.43	1.61	0.89	0.46	0.26	0.49	0.75		0.66	0.56	
<i>n</i> -C ₄	2.37	0.88	3.03		0.46	0.33	0.72	1.26	0.82	2.66	1.23	3.02
<i>i</i> -C ₅	1.06	0.31	0.60	4.36	0.26	0.16	0.34	0.41		0.62	0.52	
<i>n</i> -C ₅	0.98	0.35	0.68		0.20	0.11	0.40	0.40	0.34	0.94	0.60	1.35
C ₆	1.01	0.53	0.99	3.08	0.35	0.25	0.57	0.46	0.39	1.14	0.72	0.90
C ₇₊	11.18	4.42	2.92	2.63	5.64	0.98	4.78	3.04	1.54	3.48	3.10	5.88
molecular weight, MW _{C7+}	186	171	125	106	253	122.6	200	155.3	138.78	152.3	132	153
specific gravity, SPGR _{C7+}	0.807	0.813	0.74	0.733	0.850	0.723	0.820	0.8311	0.7961	0.7763	0.774	0.81
dew point temperature, <i>T</i> _{dew} (K)	394.82	422.04	277.59	288.71	405.93	377.59	379.82	392.04	367	363	355.6	408.7
dew point pressure, <i>P</i> _{dew} (MPa)	36.053	54.268	21.339	18.278	81.565	23.063	60.329	39.851	26.46	23.53	28.1	33.47

^a Superscripted values next to each of the 12 SNG mixtures denote the reference source.

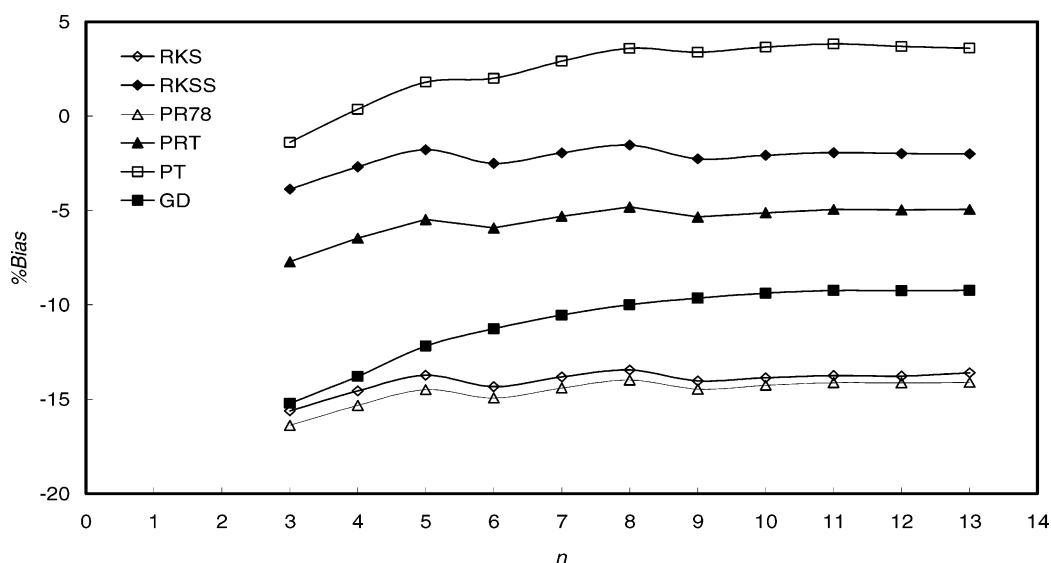


Figure 11. Deviation plot for natural gas mixture A, showing the percent deviation in predicting dew point, as a function of the number of single carbon numbers (SCNs).

for the other EoSs. However, the RKSS EoS, as a representative of the RK family, and the SW EoS describe the dew point loci significantly better than the others.

The synthetic natural gases used in this study are considerably lean in heavy hydrocarbons. A real natural gas is usually rich in heavy hydrocarbons. All heavy hydrocarbons are usually grouped together and reported as C₇₊. It is then important to split the C₇₊ fraction into several SCN groups and calculate the critical properties of each group if an EoS is to be used to calculate the natural gas dew point.

Table 7 gives the specifications of real natural gases

used in this study. Mixture A, which contains 11.18% C₇₊, is used to find the optimum number of SCN groups to calculate the natural gas dew point using different EoSs. This evaluation is given in Figure 11 in terms of %Bias:

$$\% \text{Bias} = \frac{P_{\text{d,cald}} - P_{\text{d,expl}}}{P_{\text{d,expl}}} \times 100 \quad (8)$$

Figure 11 shows that, if the C₇₊ fraction in mixture A is divided into more than 10 SCN groups, all the EoSs become stable and predict the dew point of mixture A uniformly. However, to a large extent, the prediction ability of EoSs is dependent on the critical properties of the SCN groups. This is best observed in Figure 12, where the %Bias of the GD EoS, as a representative of the EoSs, is plotted against the number of SCNs. Different correlations give different predictions; however, the correlations of Lee and Kesler²⁹ (LK) and Twu²⁸ seems to be better than those of Cavett²⁶ and Riazi and Daubert²⁷ (RD). Figure 12 also shows that the predictions by the GD EoS become improved and

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 (39) Sage, B. H.; Olds, R. H. *T. Soc. Petrol. En. AIME* **1947**, *170*, 156–173.

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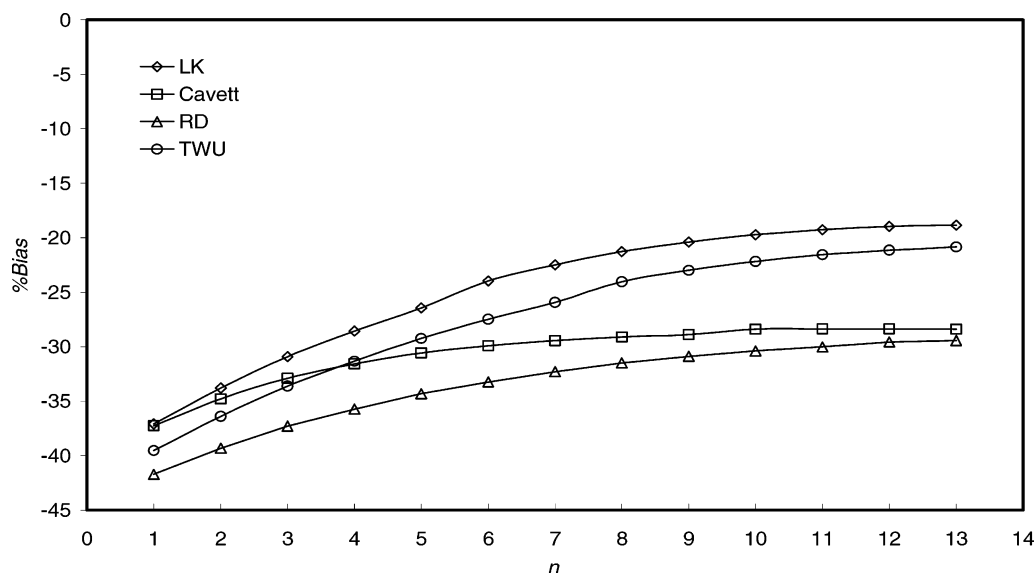


Figure 12. Deviation plot for natural gas mixture B, showing the percent deviation in predicting dew point using the GD EoS, as a function of the number of SCNs.

Table 8. Prediction of the Natural Gas Dew Point Pressures, in Terms of %AAD, from the Equations of State

SCN group	%AAD of the Predicted Dew Point Pressure												
	RKS	RKT	RKSS	PR76	PR78	PRT	PRF	PRG	PT	PTV	SW	GD	TBS
A	19.21	15.50	8.39	21.47	19.53	11.01	6.35	17.61	6.89	8.91		14.41	16.14
B	8.24	8.90	27.19	2.39	4.80	18.93	35.77	21.24	8.57	12.21	12.88	24.52	39
C	2.24	5.36	5.49	3.92	4.66	2.56	10.51	2.98	3.24	2.89	3.55	2.61	9.81
D	2.50	7.17	9.45	0.90	0.68	5.25	14.77	1.35	2.21	4.23	7.74	1.21	12.95
E	31.90	27.92	18.26	38.17	35.68	26.34	21.46	33.63	19.99	9.67		23.30	12.47
F	17.23	13.74	8.28	27.67	25.88	18.78	15.37	24.12	23.83	20.19	11.65	17.85	46.77
G	21.49	18.84	13.20	28.43	26.01	16.63	10.53	23.94	14.38	11.64	37.83	14.32	21.53
H	26.18	23.39	18.01	31.90	30.36	24.12	20.61	28.92	28.02	23.62	10.53	22.46	34.07
I	9.17	15.01	22.36	4.54	5.26	11.08	17	5.37	6.25	11.13	30.05	12.13	9.67
J	10.04	15.34	21.37	4.52	5.99	14.86	20.41	7.99	9.10	14.86	28.33	10.63	10.95
K	2.28	4.08	9.74	7.63	6.16	2.15	7.71	4.39	3.48	1.96	22.16	1.86	12.42
L	9.90	6.69	3.96	16.02	14.43	7.41	4.14	12.61	11.07	6.51	6.17	7.42	21.53
average	13.64	13.49	13.81	15.32	14.95	13.26	15.38	15.34	11.42	10.65	17.09	12.73	20.61

unchanged for 12 or more SCNs, regardless of which correlation is used to characterize the C_{7+} fraction. Considering Figures 11 and 12, we have concluded that 12 SCN groups are suitable for dew point calculations. Previously, Elsharkawy³⁸ reported a similar conclusion, having examined the dew point prediction of the RKS EoS for a natural gas mixture.

In Table 8, the %AADs in predicting the dew points of natural gases given in Table 7 are presented. In these calculations, the C_{7+} fractions were divided into 12 SCN groups. The values given in Table 8 are, in fact, the average values. Using each EoS, the dew point of each mixture has been predicted four times. Each time, one of the correlations of LK, Cavett, Twu, and RD has been used to calculate the critical properties and the Edmister⁴⁶ equation was used to calculate the acentric factor. The calculated values were then averaged and have been reported in Table 8. As can be seen, the best five EoSs are, respectively, PTV, PT, GD, PRT, and RKT. Among the two families, clearly the RK family is superior and should be preferred. It is also valuable to find the best correlations of critical properties for predicting dew points. Table 9 clearly shows that the Twu²⁸ correlation is in better agreement with the EoSs and should be used to calculate the SCN group critical properties.

Table 9. Prediction of the Natural Gas Dew Point Pressures, in Terms of %AAD, Using Different Characterization Methods

equation of state, EoS	%AAD of Dew Point Measurement			
	LK	Cavett	RD	Twu
RKS	12.99	14.58	15.36	11.62
RKT	13.27	14.53	16.01	11.05
RKSS	11.56	14.18	14.07	16.13
PR76	13.66	17.29	18.92	14.90
PR78	13.27	16.35	17.92	13.38
PRT	12.14	13.68	14.26	13.93
PRF	13.06	17.97	14.67	14.08
PRG	18.03	15.21	16.83	12.46
PT	10.37	12.57	13.46	9.28
PTV	10.97	11.35	10.91	9.51
SW	19.82	16.78	16.58	15.17
GD	10.45	14.07	15.22	11.17
TBS	21.52	21.16	25.81	18.21
average	13.93	15.36	16.16	13.15

6. Conclusions

The dew points of lean and rich natural gas mixtures have been predicted using 15 equations of state (EoSs). The supercritical behavior of methane and nitrogen, and the vapor pressure of the components forming natural gases, have been studied using these EoSs. It can be inferred that, if an EoS is capable of predicting the fugacity of methane under supercritical conditions and

(46) Edmister, W. C. *Pet. Refin.* **1958**, 37, 173–179.

the vapor pressure of natural gas components accurately, especially the heavy ones, one can expect the EoS to be accurate in regard to predicting the natural gas dew points. All the EoSs in the RK family and the GD EoS are good examples.

The impact of binary interaction parameters (BIPs) on the prediction of EoSs has been studied. It has been found that, for synthetic natural gases, the use of BIPs does not necessarily improve the dew point prediction accuracy of the EoSs. This conclusion suggests that the critical properties and acentric factor used in this work to calculate the EoS parameters may not be the same as those used for optimizing BIPs.

The lean synthetic natural gas dew points have best been predicted using the EoSs from the RK family, i.e., the RKS, RKSS, and RKT EoSs and the SW EoS. The

PRT and GD EoSs also have been determined to be promising.

It has been found that, using an exponential distribution devised by Pedersen et al.²⁵, a C₇₊ fraction must be divided into at least 12 SCN groups so that the predictions can become improved and unchanged by any of the EoSs. The use of the Twu²⁸ correlation to calculate the critical properties of SCN groups is also recommended.

Generally, it has been found that three-parameter EoSs can describe natural gases more accurately than the other EoSs. Accordingly, the PTV, PT, and GD EoSs are recommended for predicting rich natural gas dew points. For lean natural gases, however, the RKS, RKSS, RKT, and SW EoSs are superior.

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