Accurate Density and Viscosity Modeling of Nonpolar Fluids Based on the ''f-Theory'' and a Noncubic Equation of State¹

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Recently, the f-theory, a theory for viscosity modeling based on friction concepts of classical mechanics, has been introduced. This new theory allows accurate viscosity–pressure–temperature $(p-p-T)$ modeling based on a van der Waals type of equation of state, one with a repulsive pressure term and an attractive pressure term. Thus, popular cubic equations of state (CEOS), such as the SRK and the PR, have been successfully applied to obtain accurate $\eta - p$ –T models (even close to the critical region) of fluids such as *n*-alkanes, N_2 , CO_2 , etc.,andsomeof theirmixtures.However,even thoughithasbeenshownthataCEOS f-theory-based model can accurately reproduce the viscosity behavior of, at least, nonpolar fluids, the accuracy of the density predictions is still limited by the algebraic structure of the CEOS. In this work, a noncubic van der Waals type of equation of state is introduced for the accurate modeling of both the density and the viscosity behavior of selected nonpolar fluids. The achieved accuracy, for both the density and the viscosity fluid properties, is close to, or within, experimental uncertainty and applies to wide temperature and pressure ranges.

KEY WORDS: equation of state; f-theory; hydrocarbons; modeling; prediction; viscosity.

1. INTRODUCTION

Recently, Quiñones-Cisneros et al. [1] proposed the friction theory (*f-theory*) for viscosity modeling and illustrated the capability of this theory by modeling

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the viscosity of nonpolar fluids over wide ranges of temperature and pressure. The f-theory is based on friction concepts of classical mechanics and the van der Waals theory of fluids. The main difference between the f-theory and other approaches to viscosity modeling is that the viscosity of dense fluids is considered as a mechanical rather than a transport property. In the f-theory the viscosity is linked to the pressure, which is the main mechanical variable, and, by use of a simple cubic equation of state (CEOS), accurate viscosity estimations can be obtained from low to high pressures. This is achieved regardless of the accuracy of the estimated density. Based on the viscosity behavior of normal alkanes, from methane to *n*-octadecane, along with carbon dioxide and nitrogen, the principles of the f-theory were further developed into one-parameter general models [2] for some of the basic CEOS. Despite the fact that one-parameter f-theory models require only one tuning parameter, these models are capable of accurately reproducing the viscosity of a large number of nonpolar fluids and their mixtures. Furthermore, the use of a CEOS in combination with f-theory viscosity models has also been applied to the modeling and prediction of a large number of systems from light fluids such as the viscosity prediction of natural gas [3], light gases [4], carbon dioxide plus hydrocarbon mixtures [5], and hydrogen plus natural gas (hythane) mixtures [6] to more complex dense fluids such as the prediction and modeling of dense reservoir fluids [7]. However, the applications carried out so far with the f-theory pertain to CEOS with the inherent limitations that such models have—particularly concerning the modeling and prediction of the fluid volumetric properties. Therefore, it is of theoretical as well as of industrial interest to explore the extension of the f-theory approach to other kinds of models besides the CEOS. Of practical interest are models capable of a better density description as well as models that can be applied to associating fluids. Thus, the main task of this work is to establish the lines along which the f-theory can be extended to a noncubic equation of state.

As a first case study, this work approaches the issue of accurate simultaneous viscosity and density modeling, by extending the f-theory approach to the recent modification of the Benedict–Webb–Rubin [8] equation of state by Soave [9] (SBWR EOS). The SBWR EOS is capable of accurate density predictions for *n*-alkanes as large as $n-C_{20}$. Thus, in this work the SBWR EOS has also been extended to the accurate viscosity modeling for almost the same number of *n*-alkanes. However, since the SBWR is not a van der Waals type of EOS, i.e., it is not based on a repulsive and an attractive pressure term, to extend the f-theory to this kind of model, some considerations in that respect have been taken into account. In addition, a first attempt at predicting the viscosity of *n*-alkane mixtures with this kind of model is also considered in this work.

2. THE FRICTION THEORY

According to the f-theory $\lceil 1 \rceil$, the total viscosity η of dense fluids can be separated into a dilute gas term, η_0 and a friction term, η_f ,

$$
\eta = \eta_0 + \eta_f \tag{1}
$$

The dilute gas term η_0 applies at the zero pressure limit of the gas phase, and for many fluids it can be accurately estimated with simple empirical models such as the one proposed by Chung et al. [10]. This model is applicable for predicting the dilute gas viscosity of many polar and nonpolar fluids over wide ranges of temperature with an absolute average deviation (AAD) of 1.5%. In micropoise (μP) (1 $\mu P = 0.1 \mu Pa \cdot s$), the dilute gas model of Chung et al. is given by

$$
\eta_0 = 40.785 \frac{\sqrt{MWT}}{v_c^{2/3} \Omega^*} F_c \tag{2}
$$

where the following empirical equation is used to estimate the reduced collision integral:

$$
\Omega^* = \frac{1.16145}{T^{*^{0.14874}}} + \frac{0.52487}{\exp(0.77320T^*)} + \frac{2.16178}{\exp(2.43787T^*)} -6.435 \times 10^{-4} T^{*^{0.14874}} \sin(18.0323T^{*^{-0.76830}} - 7.27371)
$$
(3)

with

$$
T^* = \frac{1.2593T}{T_c} \tag{4}
$$

In Eq. (2) the critical volume v_c is given in cm³·mol⁻¹, and for nonpolar gases the F_c factor is empirically found to be

$$
F_{\rm c} = 1 - 0.2756\omega\tag{5}
$$

In the case of the friction viscosity term η_f , Quiñones-Cisneros et al. [1] proposed an analogy between the Amontons–Coulomb friction law and the van der Waals repulsive and attractive pressure terms p_r and p_a . Thus, by means of three temperature-dependent coefficients, η_f can be linked to the van der Waals repulsive and attractive pressure terms as follows:

$$
\eta_{\rm f} = \kappa_{\rm r} p_{\rm r} + \kappa_{\rm rr} p_{\rm r}^2 + \kappa_{\rm a} p_{\rm a} \tag{6}
$$

This approach, in conjunction with a simple cubic EOS, can provide accurate viscosity modeling of *n*-alkanes over wide ranges of temperature and pressure. Furthermore, by using simple mixing rules, the f-theory has also been found to give accurate mixture viscosity predictions without any need of viscosity binary parameters [1, 2].

3. THE SBWR MODEL

Although the noncubic EOS recently proposed by Soave [9] can provide accurate densities for *n*-alkanes with acentric factors as large as 0.9, f-theory viscosity modeling based on this type of EOS is not straightforward. The f-theory is based on the van der Waals concept of a balance between the repulsive and the attractive pressure terms, while a BWR type of EOS is not structured in this way. However, there are alternative ways by which the f-theory can be adapted to an EOS that is not of the van der Waals type. For example, if the total pressure in a given EOS is written as the addition of *n* terms,

$$
p = \sum_{i=1}^{n} p_i \tag{7}
$$

a straightforward extension of the f-theory is to write an expression for the friction viscosity term as follows:

$$
\eta_{\rm f} = \sum_{i=1}^{n} \left(\kappa_{i,1} p_i + \kappa_{i,2} p_i^2 \right) \tag{8}
$$

In such a case, a term-by-term analysis may show that some of the second-order terms may be neglected if they do not have an important contribution at high pressures. This approach has been tested for the SBWR EOS, and accurate modeling results have been obtained for pure components. However, if the viscosity of mixtures is predicted using the same simple mixing rules as with other f-theory models [1, 2], the results may not be as good due to the large discrepancies in the magnitude of the different SBWR EOS terms as the model is applied to different substances. Therefore, if a model such as the one given in Eq. (8) is used for the prediction of mixture viscosities, adequate mixing rules have to be developed. In addition, although a straightforward extension of the f-theory, a model such as Eq. (8) may result in an unnecessarily large number of friction parameters.

An alternative approach to accurately modeling the viscosity of pure fluids and satisfactory mixture viscosity predictions is to group the terms of

Fig. 1. The SBWR *n*-heptane reduced pressure, reduced attractive pressure term, and reduced repulsive pressure term as a function of the reduced density at a reduced temperature of 0.5.

the non-van der Waals EOS into attractive-like and repulsive-like pressure terms to obtain a model more consistent with the f-theory. In the case of the SBWR EOS, this can be achieved by separating the equation into the following attractive-like and repulsive-like pressure terms:

$$
p = p_a + p_r \tag{9}
$$

where

$$
p_{\rm a} = BRT\rho^2 \tag{10}
$$

and

$$
p_r = (1 + D\rho^4 + E\rho^2(1 + F\rho^2) \exp(-F\rho^2)) RT\rho
$$
 (11)

Figure 1 shows the p/p_c , p_a/p_c , and p_r/p_c isotherms predicted by the SBWR EOS for *n*-heptane at a reduced temperature of 0.5. Although it appears that Eqs. (10) and (11) have a correct qualitative attractive and repulsive performance, another major difference compared with a van der Waals type of EOS lies in the fact that the repulsive term is not a clearly dominating term at high pressures. This behavior is due to the lack of an excluded volume in the mathematical structure of an EOS such as the SBWR EOS. Thus, Eq. (6) cannot be directly applied since it was derived under the assumption that the repulsive term strongly dominates at high pressures. Therefore, if this assumption is removed, an extension of the f-theory to the SBWR EOS would give

$$
\eta_{\rm f} = \kappa_{\rm a} p_{\rm a} + \kappa_{\rm aa} p_{\rm a}^2 + \kappa_{\rm r} p_{\rm r} + \kappa_{\rm rr} p_{\rm r}^2 \tag{12}
$$

where κ_a , κ_r , κ_{aa} , and κ_r are the corresponding temperature-dependent linear and quadratic friction parameters.

For the temperature dependence of the friction parameters, it has been found that a regular second-order polynomial on the inverse of the reduced temperature gives accurate results. On the other hand, similarly to the quadratic term of the van der Waals f-theory models [1], an exponential term, which vanishes as the temperature increases, is required for an accurate temperature-dependent description of the second-order terms. This results in the following empirical models for the temperature dependence of the friction terms:

$$
\kappa_{\rm r} = a_0 + a_1 T_{\rm r}^{-1} + a_2 T_{\rm r}^{-2} \tag{13}
$$

$$
\kappa_{\rm a} = b_0 + b_1 T_{\rm r}^{-1} + b_2 T_{\rm r}^{-2} \tag{14}
$$

$$
\kappa_{\rm rr} = c_2 (\exp \lfloor 2T_{\rm r}^{-1} \rfloor - 1) \tag{15}
$$

and

$$
\kappa_{aa} = d_2(\exp\left[2T_r^{-1}\right] - 1) \tag{16}
$$

where

$$
T_{\rm r} = \frac{T}{T_{\rm c}}\tag{17}
$$

4. VISCOSITY MODELING OF PURE NORMAL ALKANES

To derive general models for *n*-alkanes, an overall least-squares (LS) fitting to a database of recommended viscosity data [11] has been performed for the f-theory SBWR model. This is the same database used for the one-parameter general models [2]. In addition, a LS fitting of the recommended *n*-nonane and *n*-undecane data of Stephan and Lucas [12] has also been carried out. The SBWR EOS has been used as described by Soave [9] regarding all of the model parameters and parametric laws. The critical temperature, critical pressure, and acentric factor values necessary in the SBWR EOS have been taken from the table of recommended constants of Stryjek and Vera [13], and the molecular weight values from Reid et al. [14]. For consistency, the critical volumes required in Eq. (2) have been estimated with the empirical equation for the critical compressibility used by Soave in the SBWR EOS [9, 15],

$$
Z_{\rm c} = 0.2908 - 0.099\omega + 0.04\omega^2\tag{18}
$$

The f-theory SBWR friction constants, required in Eqs. (13)–(16), have been obtained through a LS fitting of the considered *n*-alkane data, as described above, and they are listed in Table I. Table II lists the temperature and pressure ranges of the modeled data together with the absolute average deviation (AAD), the point of maximum deviation, and the maximum deviation value. The results reported in Table II show a very accurate model performance in all cases. In addition, all of the maximum deviations are also within experimental uncertainty. This follows from the fact that for all light *n*-alkanes, from methane to *n*-octane, the point of maximum deviation is close to the critical point where the viscosity derivative with respect to the pressure diverges. For all of the remaining dense hydrocarbons, the maximum deviation value is not larger than the experimental uncertainty of the original data.

Figures 2, 3, and 4 show the f-theory SBWR model results for a light alkane (propane), an intermediate one (*n*-heptane), and a dense one (*n*-pentadecane), respectively. The propane results (Fig. 2) show good agreement with

Fig. 2. Propane viscosity results $(1 \mu P = 0.1 \mu Pa \cdot s)$ for the f-theory SBWR model (solid curves) along with the data recommended by Zéberg-Mikkelsen [11] (points).

					Point of max. dev.	
	T/T_c (range)	p/p_c (range)	AAD $(\%)$	T/T_c	p/p_c	Max. dev. $(\%)$
Methane	$0.55 - 2.50$	$0 - 20.0$	0.95	1.0	1.05	6.44
Ethane	$0.35 - 1.60$	$0 - 11.3$	2.01	1.0	0.60	11.4
Propane	$0.30 - 1.29$	$0 - 13.0$	2.72	1.1	0.8	10.3
n -Butane	$0.35 - 1.04$	$0 - 18.0$	1.74	1.0	0.6	16.1
n -Pentane	$0.64 - 1.16$	$0 - 30.0$	1.14	1.1	0.8	6.78
n -Hexane	$0.54 - 1.08$	$0 - 35.0$	1.38	1.0	0.95	12.4
n -Heptane	$0.56 - 1.00$	$0 - 35.0$	0.67	1.0	0.95	8.65
n -Octane	$0.50 - 1.00$	$0 - 40.0$	1.09	1.0	0.95	7.90
n -Nonane	$0.50 - 0.79$	$0 - 30.2$	0.66	0.64	4.4	3.71
n -Decane	$0.45 - 0.76$	$0 - 48.0$	0.49	0.65	8.0	1.80
n -Undecane	$0.47 - 0.81$	$0 - 25.4$	0.45	0.81	20.3	2.98
n -Dodecane	$0.45 - 0.60$	$0 - 55.0$	0.52	0.6	54.8	1.62
n -Tridecane	$0.44 - 0.53$	$0 - 60.0$	0.35	0.525	60.0	1.25
n -Tetradecane	$0.42 - 0.54$	$0 - 60.0$	0.63	0.51	0.2	2.09
n -Pentadecane	$0.44 - 0.58$	$0 - 65.0$	0.65	0.575	65.0	2.15
n -Hexadecane	$0.41 - 0.52$	$0 - 70.0$	1.24	0.46	70.0	3.53
n -Octadecane	$0.42 - 0.55$	$0 - 80.0$	0.94	0.51	0.2	3.19

Table II. Overall Performance of the f-Theory SBWR Model for Pure *n*-Alkanes

the data. In addition, the propane predictions show a good, consistent, and stable performance, even for the low-temperature extrapolated pressure cases where calculations have been made up to 600 bar. The same performance remarks also apply for the *n*-heptane results depicted in Fig. 3, where the maximum pressure reaches 1600 bar. Finally, the *n*-pentadecane results depicted in Fig. 4 also show a good model performance over the entire data temperature and pressure ranges.

5. VISCOSITY MODELING OF NORMAL ALKANE MIXTURES

In the original f-theory article $[1]$, it was shown that some simple mixing rules for the friction parameters provide good results for the prediction of mixture viscosities. In this work it has been found that, for the f-theory SBWR, good performance is also obtained with similar mixing rules. Thus, in the case of mixtures, the viscosity is given by

$$
\eta_{\text{mx}} = \eta_{0,\text{mx}} + \eta_{\text{f},\text{mx}} \tag{19}
$$

Fig. 3. *n*-Heptane viscosity results $(1 \mu P = 0.1 \mu Pa \cdot s)$ for the f-theory SBWR model (curves) along with the data recommended by Zéberg-Mikkelsen [11] (points).

where the subscript mx indicates the corresponding mixture property. Here, the mixture dilute gas limit is calculated by

$$
\eta_{0, \max} = \exp \left[\sum_{i=1}^{n} x_i \ln(\eta_{0,i}) \right]
$$
 (20)

In all cases, subscript *i* refers to the corresponding pure component of an *n*-component mixture. For the f-theory SBWR model, the mixture friction contribution term is given by

$$
\eta_{f, \max} = \kappa_{r, \max} p_r + \kappa_{a, \max} p_a + \kappa_{rr, \max} p_r^2 + \kappa_{aa, \max} p_a^2 \tag{21}
$$

where $\kappa_{r,mx}$, $\kappa_{a,mx}$, $\kappa_{rr,mx}$, and $\kappa_{aa,mx}$ are the viscous friction parameters for the mixture. Hence, for the viscous friction parameters the following simple mixing rules have been found to deliver satisfactory results:

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$$
\kappa_{\mathbf{r},\,\mathbf{m}\mathbf{x}} = \sum_{i=1}^{n} x_i \kappa_{\mathbf{r},\,i} \tag{22}
$$

$$
\kappa_{a,\,\text{mx}} = \sum_{i=1}^{n} x_i \kappa_{a,\,i} \tag{23}
$$

$$
\kappa_{\text{rr, mx}} = \sum_{i=1}^{n} x_i \kappa_{\text{rr, }i} \tag{24}
$$

and

$$
\kappa_{\text{aa},\text{mx}} = \sum_{i=1}^{n} x_i \kappa_{\text{aa},i} \tag{25}
$$

Table III shows the AAD results for the mixture viscosity predictions of different *n*-alkane mixtures. In general, it can be appreciated that the accuracy of the mixture predictions is satisfactory. A larger deviation is

Fig. 4. *n*-Pentadecane viscosity results $(1 \mu P = 0.1 \mu Pa \cdot s)$ for the f-theory SBWR model (curves) along with the data recommended by Zéberg-Mikkelsen [11] (points).

	Ref. No.	$AAD(\%)$	Max. dev. $(\%)$
$C_1 + C_2$	[16]	3.91	14.56
$C_1 + C_3$	[17]	4.21	11.30
$C_1 + n - C_4$	$\lceil 18 \rceil$	2.42	9.78
$C_1 + n - C_6$	[19]	4.06	9.70
$C_1 + n - C_{10}$	$\lceil 20 \rceil$	15.75	19.88
$n-C_s+n-C_s$	[21]	4.56	11.91
$n - C_5 + n - C_{10}$	$\lceil 22 \rceil$	4.84	17.79
$n - C_6 + n - C_7$	$\lceil 23 \rceil$	0.91	1.52
$n - C_7 + n - C_8$	$\lceil 24 \rceil$	2.23	7.56
$n - C_7 + n - C_9$	$\lceil 23 \rceil$	0.81	2.62
$n - C_8 + n - C_{10}$	$\lceil 25 \rceil$	3.46	6.95
$n - C_{10} + n - C_{16}$	$\lceil 26 \rceil$	4.73	9.58
$n - C_5 + n - C_8 + n - C_{10}$	[27]	2.41	11.28
$n - C_{10} + n - C_{12} + n - C_{14} + n - C_{16}$	$\lceil 26 \rceil$	5.68	9.86

Table III. Overall Performance for Viscosity Predictions of Hydrocarbon Mixtures Along with the Data Sources

found for the methane $+n$ -decane system, which may be due to a combination of factors that include the uncertainty in the experimental data, the large acentric difference between the methane and the *n*-decane, and the fact that, as suggested by Soave [9], all binary interaction parameters have been set to zero. Figure 5 illustrates the prediction results for the quaternary $n - C_{10} + n - C_{12} + n - C_{14} + n - C_{16}$ mixture.

6. CONCLUSIONS

In this work the f-theory has been extended to the SBWR EOS, a Benedict–Webb–Rubin type of EOS recently proposed by Soave [9]. Thus, highly accurate viscosity modeling of *n*-alkanes up to *n*-octadecane has been achieved. In addition, the viscosity of several *n*-alkane mixtures has also been predicted with a good degree of accuracy. However, it should be pointed out that, in the case of pure *n*-alkanes, the viscosity modeling results obtained in this work are as accurate as the results obtained previously with a cubic EOS, although, in contrast to CEOS, the SBWR EOS can also achieve accurate density modeling.

In general terms, this work illustrates how an equation of state, different from to a van der Waals type of EOS, can be used to generate an accurate viscosity model. Clearly, there are many other EOS that are used in different industries; a large number of them are cubic EOS, but many others are not. Therefore, following the lines presented in this and previous

Fig. 5. Mixture viscosity predictions $(1 \mu P = 0.1 \mu Pa \cdot s)$ with the f-theory SBWR model (curves) for the $n - C_{10} + n - C_{12} + n - C_{14} + n$ *n-*C*¹⁶* quaternary system along with the data of Ducoulombier et al. [26] (points).

related studies [1, 2], it may be possible to derive f-theory models based on different available equations of state to extend their capabilities to viscosity modeling.

Finally, although the general accuracy of the mixture viscosity predictions is satisfactory, the f-theory SBWR mixture results appear to be less accurate than the results obtained with f-theory models based on the cubic EOS [1, 2]. This may be due to the better repulsive and attractive structure of the cubic EOS compared to a BWR type of EOS. Nonetheless, it may also be due to the fact that in the SBWR EOS, no binary interaction parameters were used. This appears to be a good choice for most of the mixtures studies. However, the mixtures that have shown the largest AADs are those composed of molecules with a large acentric difference such as methane $+n$ -decane. Therefore, the use of binary parameters in the SBWR EOS may result in better viscosity predictions. In fact, the most acentric mixture considered by Soave [9] was methane+*n*-pentane, and in that acentric range, the viscosity predictions of the f-theory SBWR model are rather accurate. Furthermore, if a binary parameter is used for the methane + *n*-decane mixture, the AAD reduces to 8.56% for a binary parameter of − 0.1 and reaches a 5.37% AAD minimum for a binary parameter value of about -0.2 . Thus, if the SBWR EOS is to be used for phase and viscosity modeling of mixtures with large acentric differences, binary parameters may also be required.

NOMENCLATURE

- *T* Absolute temperature
- *T*_c Critical temperature
- *p* Total pressure
- *p_c* Critical pressure
- *p*^a van der Waals attractive pressure term
- *p*^r van der Waals repulsive pressure term
- *v^c* Critical volume
- *xⁱ* Mole fraction of component *i*

Greek Letters

- η Total viscosity
- η_0 Dilute gas viscosity
- η_f Friction viscosity
- $\hat{\kappa}_a$ Linear attractive viscous friction coefficient
- κ_{aa} Quadratic attractive viscous friction coefficient
- κ . Linear repulsive viscous friction coefficient
- κ_{rr} Quadratic repulsive viscous friction coefficient
- *w* Pitzer's acentric factor

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