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Free-volume theory coupled with modified group-contribution PC-SAFT for predicting viscosities of 1-alkenes

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Abstract−A modified group-contribution PC-SAFT EoS combined with the Free-volume theory (FVT), recently proposed (DOI:10.1016/j.fluid.2019.112280) to simultaneously model the fluid phase equilibria and viscosity of fluids, is extended in this work to estimate the viscosity of 1-alkenes. Generalized correlation coefficients are proposed for the FVT triplet parameter set, which makes it possible to extrapolate the viscosity prediction of similar compounds that are not included in the fitting pool. The model is validated using a large experimental data of 1-alkenes over wide range of temperature and pressure (up to 2,500 bars). For 1-pentene to 1-triacontene, the overall average absolute deviation of the experimental liquid and vapor viscosity from those calculated by the model is of 5.37% and 1.41%, respectively, which are appropriate for most industrial applications.

Keywords: 1-Alkenes, Viscosities, mg-SAFT+FVT, Prediction

INTRODUCTION

For optimal design and operation of chemical process units, an accurate and reliable knowledge of thermodynamic properties along with viscosity of hydrocarbons over a wide range of pressure/temperature conditions is a prerequisite [1,2]. Hydrocarbon viscosity is an important property for oil and gas processes simulation. This is also one of the most significant transport properties required in heat and mass transfer calculations. The optimization of industrial processes requires reliable data; however, it is not always possible to measure experimental data over a wide range of thermodynamic conditions, particularly for unstable substances. Although experimental data are available for numerous hydrocarbons, there is still a need for a generalized estimator that is able to predict the viscosity of molecules that have not yet been investigated.

In the last decades, several different viscosity models have been developed; however, most of them are just applicable to a single phase or only validated at low pressure condition [3,4], and few models have been proposed to describe the viscosity of fluids at limited range of pressure [5,6]. Based on their strength, simplicity and accuracy, the most attractive viscosity theories could be the free volume theory (FVT) [7], friction theory (FT) [8] and expanded fluid theory-Yarranton and Satyro (EFT-YS) [9]. Semi-empirical models often require complex calculations to determine fitted parameters and the temperature/pressure application ranges are limited [9,10]. Viscosity models with the advantages of simplicity and good predictive ability by coupling the viscosity theory and an equation of state [7] are such as the FVT+SAFT [11-13] or the coupling of the Eyring's absolute rate theory and cubic EoS (PR, SRK) [6] or the fluid theory-Yarranton and Satyro+PR EoS [9] or modified fluid theory-Yarranton and Satyro coupled with SAFT+cubic EoS [10] or CP-PC-SAFT EoS [5]. Most of these models gave good agreement with experimental data for different chemical families such as alkanes, aromatics, alcohols, gases and few alkenes (pressure up to 2,500 bars). To our best knowledge, only a few authors have studied the viscosities of pure alkenes [5,6].

The main purpose of this work is to extend the mg-SAFT+FVT model's applicability to 1-alkenes and their mixtures. The mg-SAFT+FVT model allows to simultaneously predict the phase equilibria and viscosities of pure fluids and their mixtures only based on their pure compound's chemical group number. When possible, the model is compared to the similar model reported in the open literatures.

THEORY

1. Modified Group Contribution PC-SAFT (mg-SAFT)

mg-SAFT is a modification of the group-contribution concept that allows one to compute the PC-SAFT EoS parameters of real fluid based on its number of functional groups that appear in the molecule. The equation of state combined with the modified group contribution approach in this paper is the original PC-SAFT [14], which could be expressed as the residual Helmholtz energy sum:

$$a^{res}=a^{hc}+a^{disp} \qquad (1)$$

where the first term (a^{hc}) is connected with the contribution from the formation of chains from hard spheres, the second term (a^{disp}) is the dispersion part of the Helmholtz energy responsible for the attractive interactions. For non-associative molecules such as 1-alkenes, PC-SAFT EoS requires three parameters (dispersive energy -

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ε/k, segment diameter - σ and segment number - m). The readers are referred directly to the original papers for more details.

In mg-SAFT, the PC-SAFT EoS parameters (ε/k, σ, m) are calculated via modified group contribution relations [15]:

$$\varepsilon = \sum_{i=1}^{n_{groups}} \sqrt[n_i]{\left(\prod_{i=1}^{n_{groups}} \varepsilon_i^{n_i}\right)} - n_{groups}\alpha_{groups} \tag{2}$$

$$\sigma = \sum_{i=1}^{n_{groups}} n_i \alpha_i \Big/ \sum_{i=1}^{n_{groups}} n_i \tag{3}$$

$$m = \sum_{i=1}^{n_{groups}} n_i m_i \tag{4}$$

In Eqs. (2), (3), (4), n_i is the number of groups i in the molecule, α_{groups} is the group's dispersive energy correction factor which retrieved by previous work (α_{groups}=0.233 K) [15].

1. Free-Volume Theory (FVT)

The FVT model used in this work was proposed by NguyenHuynh et al. [16], which is based on the free-volume concept and the assumption that viscosity of real fluids could be directly related to PC-SAFT EoS molecular parameters. The PC-SAFT molecular parameters of 1-alkenes are estimated via the modified group-contribution approach (using Eqs. (2), (3), (4)). mg-SAFT+FVT has been successfully applied to simultaneously model the fluid phase equilibria and viscosities of alkanes, aromatics, gases [16], alcohols [17,18] and their mixtures. In the equation below, the viscosity of fluids is given in mPas and consists of two terms:

$$\eta = \eta_0 + \Delta\eta \tag{5}$$

The first term, dilute gas viscosity (η_0), is expressed as [16]:

$$\eta_0 = 26.69\frac{\sqrt{MT}}{\sigma^2 \Omega^*}\frac{F_c}{10^6\, m^{1.5}} \tag{6}$$

in Eq. (6), T, M, η_0 are the temperature (K), mass molecular (g/mol) and the gas viscosity, respectively; σ and m are the PC-SAFT EoS hard-sphere diameter (Å) and segment number. For a chemical series (such as 1-alkenes), the empirical factor, F_c is defined as a function of chain length parameter of the fluid [16,17]. The reduced collision integral (Ω^*) is calculated using the following equation [19]:

$$\Omega^* = \frac{1.16145}{T^{*0.14874}} + \frac{0.52487}{\exp(0.7732T^*)} + \frac{2.16178}{\exp(2.43787T^*)} - 6.435*10^{-4}T^{*0.14874}\sin[18.0323T^{*-0.7683} - 7.27371] \tag{7}$$

The dimensionless temperature (T^*) is a function of temperature and PC-SAFT dispersive energy of pure compound [16]: $T^* = 0.4\frac{T}{\varepsilon/k}$.

The residual viscosity ($\Delta\eta$) in Eq. (5) could be estimated based on the expression previously proposed [16,17]:

$$\eta = \eta_0 + 10^{-7}\rho\, L\, E\sqrt{\frac{1{,}000}{3mRT}}\exp\left[2.10^{-5}\left(\frac{E}{RT}\right)^{1.5}\right]$$
$$\text{and } E = \alpha\rho + F_p\frac{P}{\rho}10^5 \tag{8}$$

R is universal gas constant (8.314 J/mol.K) and P is pressure (in bar). The only property yielded by the mg-SAFT is the liquid density (ρ, in kg/m^3). The three characteristic parameters of fluid according to FVT theory are the length parameter (L, in Å), is related to the molecular size, the barrier energy required for self-diffusion (α, in J m^3/(mol·Kg) and the free-volume overlap (F_p). The FVT parameter triplet set and the F_c can be determined by fitting to the experimental viscosity data.

The mg-SAFT+FVT model is extended to the mixtures of similar compounds (such as 1-alkene+1-alkene) by using the two-body approach mixing rule [16]. This mixing rule has been successfully applied to predict the viscosity of several mixtures. The equation takes the form [16]:

$$\eta_{mix} = \exp\left[\sum_i x_i \ln(\eta_i)\right] + \frac{(n_c - 1)}{100}\sum_{i\neq j}^{n_c} x_i x_j \left(\frac{1}{2}\left(\frac{\eta_i^2}{\eta_j} + \frac{\eta_j^2}{\eta_i}\right)\right)^{1.5} \tag{9}$$

n_c is number of compounds in the mixtures (e.g. binary, n_c=2, ternary n_c=3, …). η_i, η_j and x_i, x_j are liquid viscosity and molar fraction of the molecule i, j.

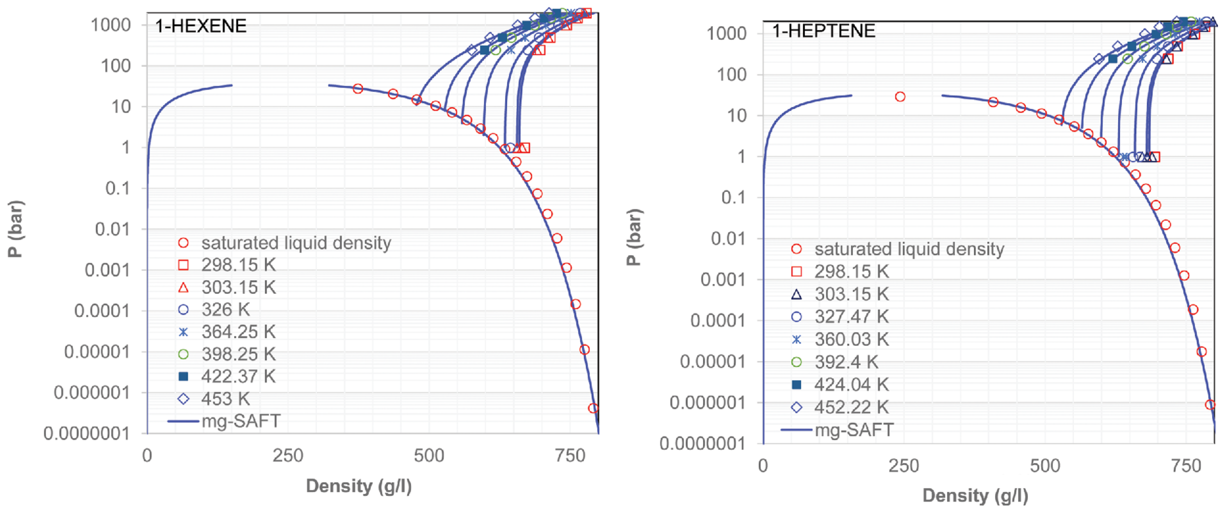

Fig. 1. Single phase densities of 1-alkenes. Symbols are experimental data [33]; solid lines are prediction results of mg-SAFT.

RESULTS AND DISCUSSION

1. mg-SAFT Parameters of 1-Alkenes

In this paper, all mg-SAFT parameters for 1-alkenes determined previously are reused without any further regression (Table 1) [20]. 1-Alkenes are composed of three types of chemical group (CH_3), (CH_2) and (CH_2=) [21,22]. Since liquid density is used as an input parameter to estimate the viscosity of 1-alkenes, the single phase liquid density of 1-alkenes was firstly predicted over wide range of temperature and pressure (up to 2,500 bars). The average absolute deviation is about 7% compared to experimental data (Fig. 1).

2. FVT Triplet Parameter Set Regression

As observed in the cases of n-alkanes [16], the incertitude of available experimental data on viscosity for 1-alkenes is typically within 1-10% [23-26]. So, only viscosity data accepted and recommended by DIPPR are used into the regression. To attach predictive character to mg-SAFT+FVT, the FVT triplet parameter set and the Fc parameter were generalized as functions of the PC-SAFT molecular parameters.

Table 1. mg-SAFT parameters for 1-alkenes [15,20]. The dispersive energy group correction factor, α_{groups}=0.223

Chemical series	Group	ε/k (K)	σ (Å)	m
n-Alkanes	(-CH_2-)	263.0678	3.9388	0.3862
	(CH_3-)	192.5045	3.4965	0.7713
1-Alkenes	(CH_2=)	243.1433	3.8210	0.4728

Based on the same spirit previously done for the alkanes [16] or alcohols [15], the smoothed experimental viscosity data over the whole ranges of temperature available in DIPPR of six alkenes from 1-hexene to 1-hexadecene were selected [27,28]. The regression of the mg-SAFT+FVT model's parameters was carried out in a sequential manner with alternate optimization of the correction factor (F_c), in Eq. (6) and then the universal constants in Eq. (8) by minimizing a quadratic residual defined by relative viscosities.

Step 1: the correction factor (F_c) of 1-alkenes is fitted using the dilute gas viscosity data. This parameter could be correlated as a function of the PC-SAFT molecular chain length parameter (m):

$$F_c=28.31596*m+188.5935 \tag{10}$$

Step 2: the FVT triplet parameter set (L, α, F_p) in Eq. (8) was obtained by regressing to the saturated liquid viscosity. These parameters are related to the PC-SAFT molecular chain length parame-

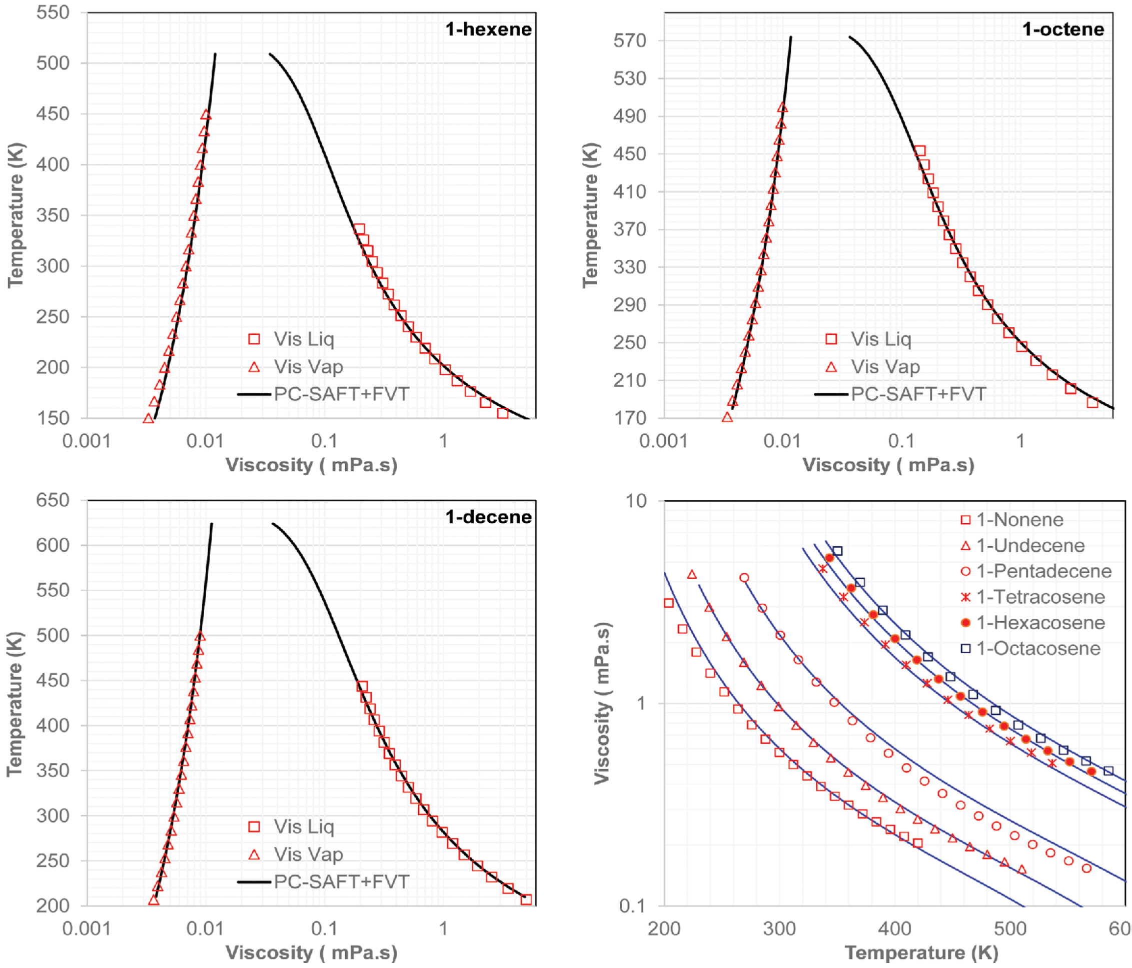

Fig. 2. Viscosities of several 1-alkenes, along the coexistence curve. Symbols are experimental data, taken from DIPPR [23]. Lines are calculated results obtained with mg-SAFT+FVT.

Table 2. The AAD for the mg-SAFT+FVT model for 1-alkenes. Experimental data are taken from DIPPR [23]

Compound	Liquid viscosity			Vapor viscosity		
	T (K)	AAD (%)	Npt	T (K)	AAD (%)	Npt
Correlation						
1-Hexene	181-335	4.70	39	181-509	4.59	82
1-Octene	220-452	4.17	58	180-574	1.37	99
1-Decene	210-442	2.84	58	210-624	1.11	104
1-Dodecene	240-524	2.44	71	240-664	1.03	106
1-Tetradecene	270-552	7.70	71	270-698	0.82	107
1-Hexadecene	280-400	6.02	30	280-724	0.99	111
OAAD		**4.34**			**1.49**	
Prediction						
1-Pentene	160-302	10.78	36	160-470	7.91	78
1-Heptene	250-400	5.83	38	180-544	1.74	91
1-Nonene	250-420	3.63	43	200-600	1.10	100
1-Undecene	230-510	2.29	70	230-644	1.06	104
1-Tridecene	260-538	4.74	70	260-682	0.88	106
1-Pentadecene	270-400	4.86	33	270-712	1.08	111
1-Heptadecene	290-400	6.69	28	290-738	1.08	112
1-Octadecene	300-400	7.72	25	300-748	1.06	112
1-Nonadecene	300-400	8.65	25	300-760	1.07	115
1-Docosene	320-540	5.95	55	320-786	0.73	117
1-Tetracosene	320-600	5.51	70	320-802	0.65	121
1-Hexacosene	330-600	3.21	68	330-816	0.66	122
1-Octacosene	340-620	3.92	70	340-828	0.77	122
1-Triacontene	340-720	11.07	95	340-840	1.05	125
1-Tetracontene				340-840	3.49	125
OAAD		**5.89**			**1.49**	

The deviation is defined as: $\mathrm{AAD}(\%)=100\cdot\sum_{data}\frac{|\eta_{exp}-\eta_{cal}|}{\eta_{exp}}$

ter (m) as follows:

$$\alpha=3695.244\mathrm{Ln}(1.17939\,\mathrm{m}) \tag{11}$$

$$\mathrm{F}_p=0.9742\mathrm{Ln}(\mathrm{m})+2.7477 \tag{12}$$

$$\mathrm{L}=177.553*10^{-5}\,\mathrm{m}+1166.753*10^{-4} \tag{13}$$

With the FVT triplet parameter set, the proposed model could reproduce well the viscosity data for most compounds in a wide range of temperature. As reported in Table 2, it is apparent that the mg-SAFT+FVT results are accurate enough for the representation of the experimental data. The overall average deviation obtained on dilute gas viscosity and saturated liquid viscosity is within the experimental accuracy (1.49% and 4.34%, respectively).

Fig. 2 illustrates the calculation of mg-SAFT+FVT model for saturated viscosities of several 1-alkenes. As it appears from this figure, our model seems to give good computation results for almost considered molecules, up to 1-octacosene.

3. Viscosity Prediction

An important factor defining the reliability of viscosity models is their extrapolative capability, particularly their performance at the extreme temperature/pressure conditions. Thus, the capability of the model by implementing the correlations (Eqs. (10) to (13)) to predict the viscosity of similar compounds is particularly important in engineering practice.

With mg-SAFT model, the only input required for 1-alkenes to estimate the PC-SAFT molecular parameters is the number of (CH_3), (CH_2) and (CH_2=) groups that appear in the molecule. For 1-alkenes that are heavier than 1-hexadecene, mg-SAFT+FVT parameters are fully extrapolated outside of the correlating range. The relative deviations obtained on vapor and liquid viscosity for heavy alkenes are presented in Table 2. From this table, the model provides generally reasonable and comparable accuracy with other thermodynamic properties obtained previously, such as vapor pressure or enthalpy of vaporization [18,28-30]. The overall agreement between calculated and experimental data is 5.89% for liquid viscosity, which is slightly higher than the correlation result; however, the accuracy of experimental data for these compounds which is lower than 10% (as recommended by DIPPR) should also be noted. Fig. 2 demonstrates the prediction results through the mg-SAFT+FVT model along with the experimental data of saturated viscosities for several 1-alkenes. As observed in the most considered cases, mg-SAFT+

FVT model can represent the experimental saturated viscosities within experimental incertitude.

The predictive ability of mg-SAFT+FVT model is then verified in a second step by applying to high pressure. This extrapolation test seems to be more stringent than in comparison with their correlation accuracy. This is demonstrated in Fig. 3, where the predic-

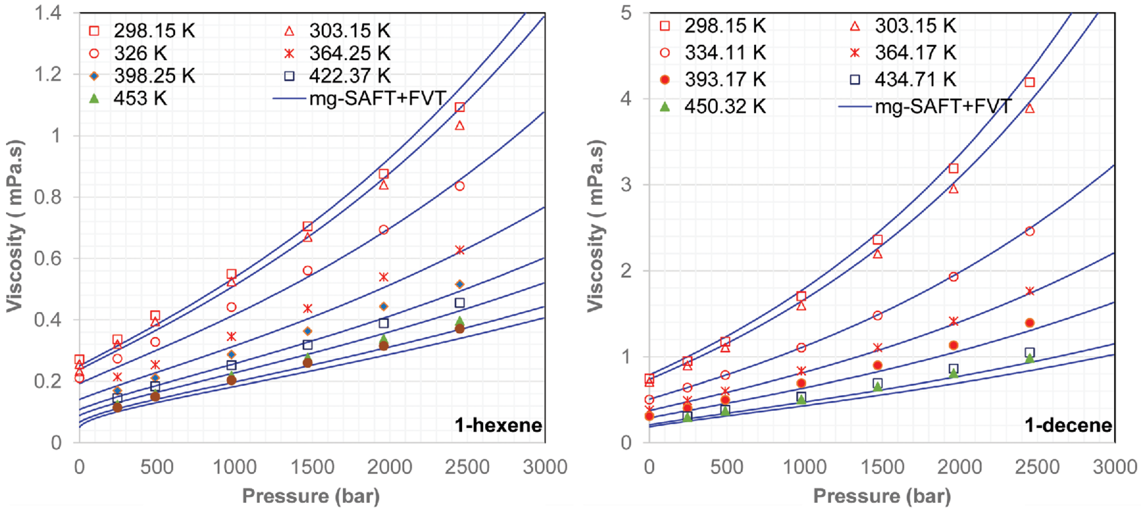

Fig. 3. Viscosities of pure 1-alkenes. Symbols are experimental data [33,34]. Solid lines: mg-SAFT+FVT prediction results.

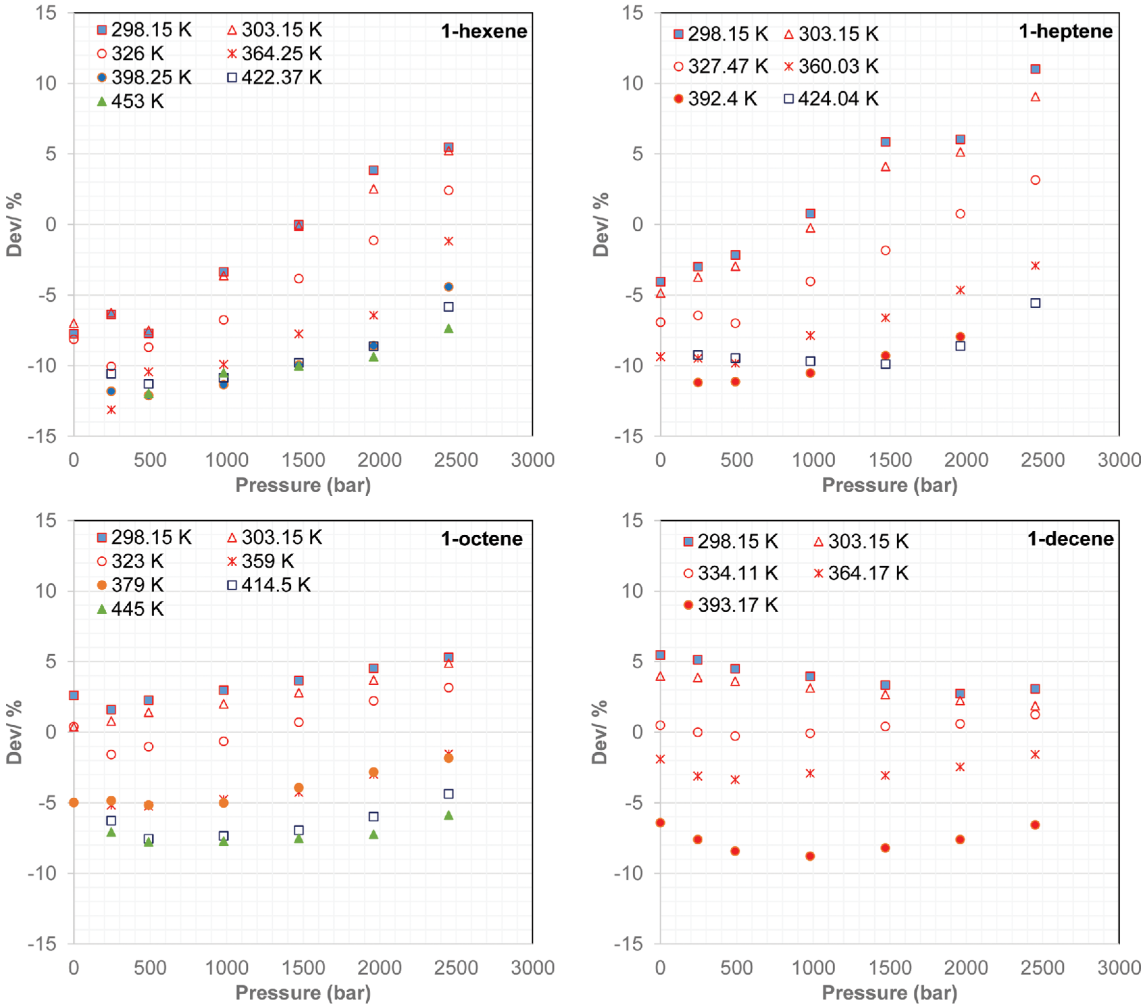

Fig. 4. Relative deviations of the computation results obtained with mg-SAFT+FVT against experimental viscosities data for pure 1-alkenes.

tions of mg-SAFT+FVT are compared with experimental data for the single phase liquid viscosity of 1-alkenes under high pressure conditions. As seen from Fig. 3, a good extrapolation to high pressure of our model was possible. The AAD results from experimental data are around 1-10%; however, for some data series at temperature higher than 450 K, this deviation is up to 15%.

The deviation between the predicted liquid viscosity of 1-alkenes obtained with the current model and the experimental data is exposed in the range of 298-475 K and up to 2,500 bars. Fig. 4 shows the relative deviations of the mg-SAFT+FVT model from the experi-

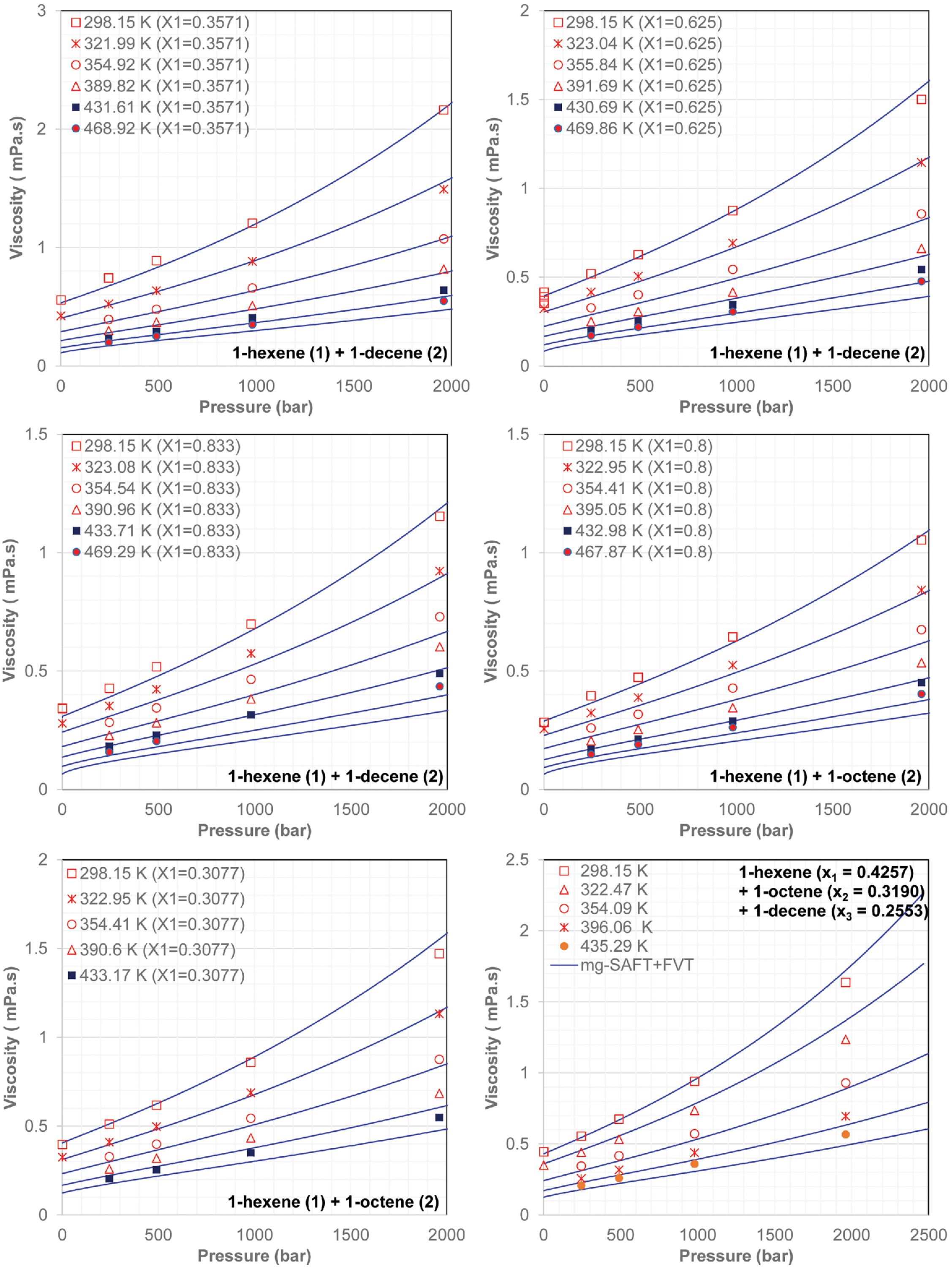

Fig. 5. Viscosities of mixtures of 1-alkene. Symbols are experimental data [24-26]. Solid lines: mg-SAFT+FVT prediction results. x_i is the mole fraction.

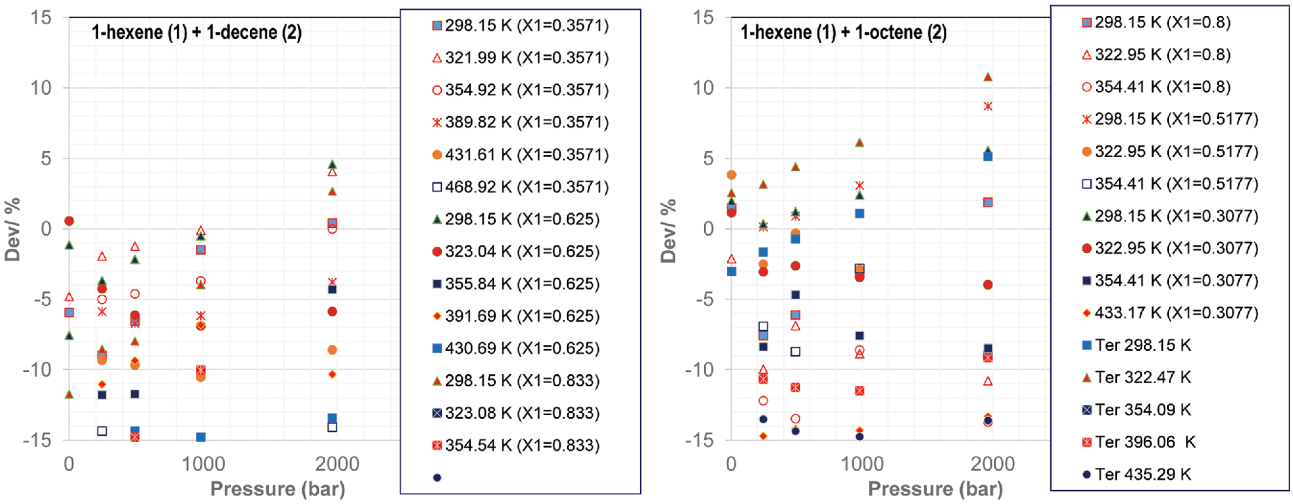

Fig. 6. Relative deviations of the computation results obtained with mg-SAFT+FVT against experimental data for the viscosities of mixtures of 1-alkene. x_i is the mole fraction. *Ter* is ternary mixture of 1-hexene (x_1=0.4257)+1-octene (x_2=0.3190)+1-decene (x_3=0.2553).

mental viscosity data for 1-hexene, 1-heptene, 1-octene and 1-decene at pressures up to 2,500 bars for different temperatures. The accuracy of the present model in calculating the high pressure viscosity at temperature lower than 450 K is observed to be better to that at above 450 K. The model systematically underestimated high pressure liquid viscosity of 1-alkenes (up to −15% compared to experimental data). The computation results show that this high deviation was not due to the accuracy of the model in calculating the viscosity of pure 1-alkene at atmospheric pressure; this reflects a temperature-dependent requirement of F_p parameter. Note that several authors measured the viscosity of 1-alkenes with a quoted uncertainty of 1 to 10% [23-26]; for the data under pressure, this uncertainty is higher.

The final step to assess the predictive capability of the mg-SAFT+FVT model is extending to the mixtures. The extension to mixtures of mg-SAFT+FVT has been detailed in ref. [17]. The mixing rule described in Eq. (9) was used in this section since all considered mixtures are similar molecules containing systems. The liquid viscosity of 1-alkene+1-alkene mixtures is calculated in a fully predictive manner, directly from viscosity of pure components without using any corrective interaction parameters. Recall that for 1-alkene containing mixtures considered in this section, only the number of (CH_2), (CH_3) or (CH_2=) groups appearing in the molecule are required as input parameters.

The experimental data on liquid viscosity of alkene containing mixtures are available only for a few binaries. To evaluate the model, we selected viscosity data of two binary and one ternary mixtures that the measured data are available over a wide range of thermodynamic conditions. For illustration, the viscosity diagrams of 1-hexene+1-octene, 1-hexene+1-decene and 1-hexene+1-octene+1-decene mixtures are depicted in pressure versus mixture's viscosity at several molar fractions (Fig. 5). It has been shown that our model yields similar deviation in comparison with the single phase high pressure viscosity data of pure compound, and it can be recognized as satisfactorily accurate, with average absolute deviation about 3 to 15% compared to experimental data.

The deviation of the present model from the experimental viscosity data for these systems as a function of pressure for different temperatures and compositions is shown in Fig. 6. The present model has similar accuracy in estimating the mixtures' viscosity compared to that obtained on pure fluids. Based on the results obtained in this section, it could be confirmed that mg-SAFT+FVT provides satisfactory prediction of mixtures' viscosity in a wide range of conditions.

4. Comparison with other Models

A direct comparison of our approach with other similar viscosity models (using an equation of state coupled with a viscosity theory) is not feasible, since the calculation with different models did not use the same experimental data points and sources; moreover, most of the published models do not report the deviation for each case. To the best of our knowledge, only a few authors attempted to propose the models predicting the viscosity of 1-alkenes and their mixtures in a systematic manner. Also, among these viscosity models, some studies have been only applied to pure 1-alkenes and not to their mixtures [5,6]. We cite here some other empirical models in which our approach results in the same level of accuracy or slightly higher deviation compared to experimental data in representing the saturated liquid viscosity of 1-alkenes. For example, Hashim et al. [31] proposed an empirical rule to correlate the viscosity of 1-alkenes. Gürbüz Yücel et al. [32] implemented Enskog's hard sphere theory to model the viscosity of 1-alkenes at atmospheric pressures. Note that our model was developed on a wide range of temperature and applied to high pressure condition.

The performance of mg-SAFT+FVT in predicting high pressure liquid viscosity of pure alkenes was compared to that obtained with CP-PC-SAFT+Modified Yarranton-Satyro [5] and Eyring+SRK [6] models. As graphically shown in Fig. 7, in all considered cases, a slightly more accurate model was observed with mg-SAFT+FVT. Through the above comparison results, our model has demonstrated the ability to correlate/predict viscosities of 1-alkenes with a very satisfactory accuracy; it can be a reliable viscosity model for industrial purposes.

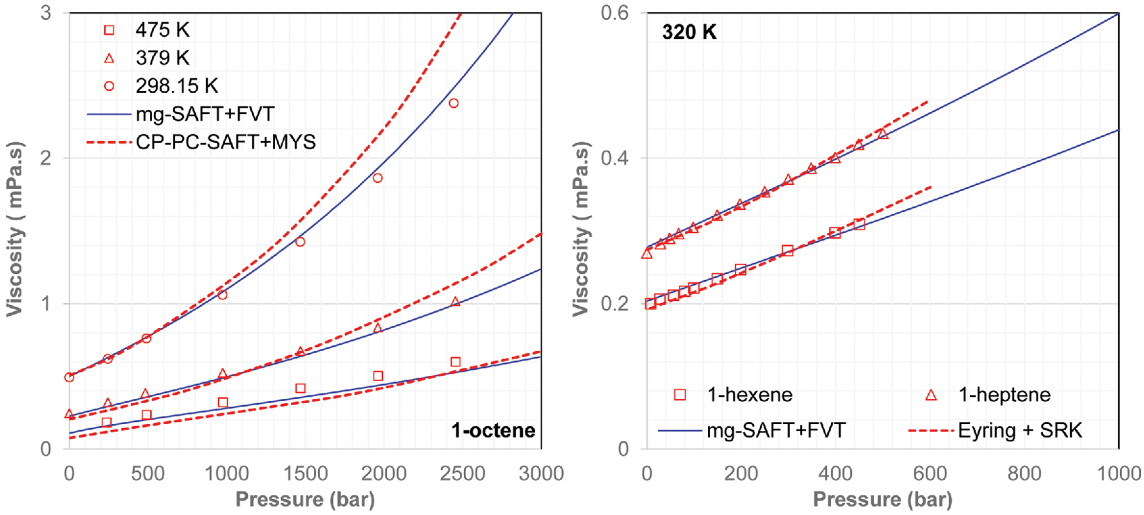

Fig. 7. Viscosities of 1-alkenes molecules. Symbols are experimental data [35]. Solid lines: mg-SAFT+FVT calculation. Dash red lines calculation of CP-PC-SAFT+MYS [5] and Eyring+SRK [6] models.

CONCLUSION

The capability of mg-SAFT+FVT model was evaluated to model the viscosity of 1-alkene and their mixtures. Our model provides a good correlation and prediction of the viscosity of this chemical series. Consequently, it can be used with confidence to predict the viscosity of these compounds under other conditions of temperature and pressure. The results obtained in the current study demonstrated that:

- For 1-alkenes, our model requires only the number of (CH_3), (CH_2) and (CH_2=) groups appearing in the molecule as input information, which allows to fully estimate the mg-SAFT+FVT parameters.

- The finding in the current paper clearly indicates that mg-SAFT+FVT could be used as an estimator to calculate the viscosity of several 1-alkenes over a wide range of temperature and pressure with average absolute deviation within the experimental incertitude.

SYMBOLS

AAD : average absolute deviation
CP-PC-SAFT : critical point-based modified PC-SAFT
DIPPR : design institute for physical property data
EoS : equation of state
mg-SAFT : modified group-contribution PC-SAFT
FVT : free-volume theory
FT : friction theory
EFT-YS : expanded fluid theory-Yarranton and Satyro
Npt : number of data points
P : pressure [bar]
PC-SAFT : perturbed-chain statistical associating fluid theory
PR : peng-robinson
SAFT : statistical associating fluid theory
SRK : Soave-Redlich-Kwong
T : temperature [K]

Subscripts

exp : experimental
calc : calculated

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