

Modeling the Viscosity Versus Temperature and Pressure of Light Hydrocarbon Solvents

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Our viscosity-temperature correlation (Miadonye and D'Orsay, *J. Mater. Eng. Perform.*, 2006, 15(1), p 13-18; Ref 1) has been extended in this work to estimate the viscosity of light hydrocarbons between a temperature range of 223.15 and 433.15 K and a pressure range from 0.1 to 240 MPa. The correlation was modified to include a pressure term that contains the pressure parameters Ψ and Ω . The value for Ω was obtained as a constant, and the value of the parameter Ψ was derived from viscosity-temperature-pressure relationship and is unique for each hydrocarbon sample. With the pressure parameter, Ψ , and the shape factor constant, Φ , the model produces an average absolute deviation of 2.3% for a total number of 503 data points, an improvement of 46% on correlations with similar simple characteristics. The value of Ψ has been determined for several light hydrocarbon samples, including crude oil fractions.

Keywords correlation equation, crude oil fractions, light hydrocarbon viscosity, pressure, solvents, temperature, viscosity model

1. Introduction

In the ever-growing oil and gas industry, the ability to predict the viscosity of hydrocarbons over a large range of conditions with reliable accuracy is of major importance. Viscosity data is essential to complete the necessary calculations used to design a variety of processes, equipment, and transportation systems. Much research has been done on formulating theoretical models designed to estimate the viscosity of substances (Ref 2-10). However, several models are inefficient in their estimating abilities and can cause large prediction error when used over a wide range of temperatures or when used on substances other than the one from which the model was correlated. Thus, a more versatile and accurate prediction model is needed.

Comuñas et al. (Ref 4) proposed an empirical equation that included the Andrade equation and a Tait-like equation for prediction of the viscosity of liquids with low molecular weights as a function of temperature (Kelvin) and pressure (MPa). The correlation has seven adjustable parameters, as shown in Eq 1 and 2:

$$\eta(P,T) = A \exp\left[\frac{B}{T-C}\right] \exp\left[D \cdot \ln\left(\frac{P+E(T)}{0.1+E(T)}\right)\right] \quad (\text{Eq 1})$$

where

$$E(T) = E_0 + E_1 + E_2 T^2 \quad (\text{Eq 2})$$

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The parameters have to be determined for each set of samples and require numerous, cumbersome calculations. Although it produced an average absolute deviation (AAD) of 0.60% for dimethyl carbonate, 0.90% for TriEGDME, 1.00% for diethyl carbonate, and 1.30% for TEGDME, the cost of cumbersome computation puts this model at a disadvantage.

Boned et al. (Ref 5) proposed a generalized model known as the self-referencing model. It requires one experimentally determined measurement at a reference temperature of 273.15 K (T_0) and atmospheric pressure, and does not require any other physical properties such as molecular weight, molar volume, or density. The self-referencing model is given as follows:

$$\ln\left(\frac{\eta(P,T)}{\eta(0.1 \text{ MPa}, T_0)}\right) = (ay^2 + by + c) \ln\left(1 + \frac{P - 0.1}{dy^2 + ey + f}\right) + (gy_0^2 + hy_0 + i) \left(\frac{1}{T} - \frac{1}{T_0}\right) \quad (\text{Eq 3})$$

where

$$y = y_0 + (gy_0^2 + hy_0 + i) \left(\frac{1}{T} - \frac{1}{T_0}\right) \quad (\text{Eq 4})$$

and

$$y_0 = \ln[\eta(0.1 \text{ MPa}, T_0)] \quad (\text{Eq 5})$$

In this correlation, T is temperature (Kelvin) and P is pressure (MPa). This model consists of nine constants ($a, b, c, d, e, f, g, h,$ and i) that are unique for each hydrocarbon sample. This correlation produced an AAD of 5.00% on a ternary mixture and 3.60% on a quinary mixture. The self-referencing model was also tested on methylcyclohexane, *cis*-decalin, and 2,2,4,4,6,8,8-heptabemethylnonane in Zéberg-Mikkelsen (Ref 6). This produced an AAD of 0.66% for methylcyclohexane, 0.99% for *cis*-decalin, and 1.45% for 2,2,4,4,6,8,8-

heptabemethylnonane. The disadvantage of this correlation is the large number of coefficients required to complete the calculations. This reduces proficiency and increases computation cost. Also, the self-referencing model's accuracy decreases considerably at low temperatures and at high pressures.

Another model, referred to as the free-volume viscosity model, was discussed in Zéberg-Mikkelsen (Ref 6). This pressure and temperature function model produced an AAD of 1.24% for methylcyclohexane, 1.22% for *cis*-decalin, and 2.05% for 2,2,4,4,6,8,8-heptabemethylnonane. The general expression used is represented by Eq 6 and 7:

$$\eta = \eta_0 + p\ell \left(\frac{\alpha\rho + \left(\frac{PM_w}{\rho}\right)}{\sqrt{3RTM_w}} \right) \times \exp \left[B \left(\frac{\alpha\rho + \left(\frac{PM_w}{\rho}\right)^{\frac{3}{2}}}{RT} \right) \right] \quad (\text{Eq 6})$$

where

$$\ell = \frac{L^2}{b_f} \quad (\text{Eq 7})$$

The variables η , η_0 , α , ρ , ℓ , M_w , R , B , L^2 , and b_f represent dynamic viscosity (Pa · s), dilute gas viscosity, characteristic parameter, density, characteristic molecular length, molecular weight, gas constant, characteristic of the free-volume overlap, average characteristic molecular quadratic length, and dissipative length, respectively. These variables are measured in SI units and are unique to every hydrocarbon. The variables α and B are parameters that need to be determined, while the remaining variables have to be measured experimentally.

In a recent publication, Xuan et al. (Ref 7) proposed a correlation to predict the viscosity of Newtonian liquids. The model, which is based on an equation of state, produces average deviations of 0.76%. The model, as given in Eq 8-14, contains two parameters that are determined from a combination of numerous physical constants:

$$\mu = \kappa_1 \exp(\kappa_2 Z) \quad (\text{Eq 8})$$

where

$$Z = \frac{1 + a\eta + b\eta^2 - c\eta^3}{(1 - \eta)^3} + r \sum_m^9 \sum_n^4 m A_{mn} \left(\frac{3\sqrt{2}}{\pi} \right)^m \eta^m \tilde{T}^{-n} \quad (\text{Eq 9})$$

$$\tilde{T} = k \left(\frac{T}{\varepsilon} \right) \quad (\text{Eq 10})$$

$$\eta = \frac{\pi r \sigma^3}{6V} \quad (\text{Eq 11})$$

$$a = r \left(1 + \frac{r-1}{r} a_2 + \frac{r-1}{r} \frac{r-2}{r} a_3 \right) \quad (\text{Eq 12})$$

$$b = r \left(1 + \frac{r-1}{r} b_2 + \frac{r-1}{r} \frac{r-2}{r} b_3 \right) \quad (\text{Eq 13})$$

and

$$c = r \left(1 + \frac{r-1}{r} c_2 + \frac{r-1}{r} \frac{r-2}{r} c_3 \right) \quad (\text{Eq 14})$$

The variables μ , κ_1 , κ_2 , Z , A_{mn} , \tilde{T} , ε , η , r , and σ are dynamic viscosity (MPa/s), temperature parameters one and two, compressibility factor of liquid, numerical coefficient, reduced temperature, attractive energy, reduced density, and collision diameter, respectively. Not only are the parameters κ_1 and κ_2 unique for each liquid, but they are also unique at different temperatures for each liquid. Therefore these parameters have to be determined for numerous temperatures for each liquid. The variables a_i , b_i , and c_i ($i = 2, 3$), are universal constants. This correlation is complicated and time consuming.

2. Development of Viscosity-Pressure Correlation

The correlation proposed by Puttagunta et al. (Ref 2) is based on the characteristics that relate kinematic viscosity of the oil with changes in temperature. It is one of a few viscosity models in the petroleum industry that is simple to use for modeling the viscosities of conventional crude oil and light hydrocarbons. The model has already been shown to be capable of delivering high-accuracy viscosity modeling of crude oils based on one measurement of viscosity at a reference temperature and 1 atm pressure (Ref 11-13). The correlation, given in Eq 15, was originally developed using 65 data sets from 20 different crude oils recovered from various locations around the world. The result obtained was excellent, with an overall AAD of 0.82% for a total of 181 individual data points:

$$\log \eta = \frac{b}{1 + \left(\frac{T - 37.78}{310.93} \right)^S} + C \quad (\text{Eq 15})$$

where η is kinematic viscosity (cSt), T is temperature ($^{\circ}\text{C}$), C is given as -0.8690 , b is the characterization parameter defined as $b = \log_{10} \eta_{37.78\text{ }^{\circ}\text{C}} - C$, and S is the shape factor, which relates to the characterization parameter as follows: $S = 0.28008b - 1.6180$.

In an attempt to develop the most accurate and simple to use model for η - T - P relationship for light hydrocarbon liquids, the correlation of Puttagunta et al. (Ref 2) was modified to incorporate a pressure term that was originally defined in the viscosity model proposed in several earlier publications (Ref 14-17) for predicting the viscosity of bitumen and heavy oils. This correlation is based on variables that relate viscosity to temperature and pressure. The modified correlation is given in Eq 16.

$$\ln \eta = 2.3026 \left[\frac{b}{1 + \left(\frac{T - 37.78\text{ }^{\circ}\text{C}}{310.93\text{ }^{\circ}\text{C}} \right)^S} + C \right] + B_0 P \exp(dT) \quad (\text{Eq 16})$$

In Eq 16, P is gauge pressure (MPa), T is temperature ($^{\circ}\text{C}$); the values for the pressure parameters B_0 and d were obtained by nonlinear regression of viscosity-pressure values, and are given as follows:

$$B_0 = 0.002889571b + 0.000947732 \quad (\text{Eq 17})$$

and

$$d = 0.005914526b - 0.008331984 \quad (\text{Eq 18})$$

The variable b is the characterization parameter that is unique to each individual hydrocarbon. For the determination of b , it is necessary to measure the viscosity of the light hydrocarbon in question at the reference temperature and one atmosphere pressure. This is the only parameter that requires any experimental measurements to compute the viscosity of that hydrocarbon at any temperature and/or at any pressure.

This model yields an AAD of 4.77% when tested on 15 different hydrocarbons, which include toluene, *cis*-decalin, decane, dimethyl carbonate, diethyl carbonate, TriEGDME, TEGDME, and several cuts from Arabian light crude oil. This deviation is too high to be considered accurate. For more accurate viscosity predictions for the 15 solvents, the S temperature parameter, and the B_0 and d pressure parameters can be

Table 1 Pressure compressibility factor (Ψ) and viscosity reduction factor (Φ) values for different solvent group

Sample	Ψ value	Φ value(a)
Toluene	0.0026920	1.9283
<i>cis</i> -Decalin	0.0048250	1.8670
Decane	0.0047193	1.8727
Dimethyl carbonate	0.0032985	2.4560
Diethyl carbonate	0.0036373	2.1906
TriEGDME	0.0033754	2.1066
TEGDME	0.0033806	2.0960
Arabian Light Cut 150-162.5 °C	0.0041898	1.9239
Arabian Light Cut 162.5-185 °C	0.0048670	1.9239
Arabian Light Cut 185-206.1 °C	0.0043832	1.9239
Arabian Light Cut 206.1-225.9 °C	0.0049897	1.9239
Arabian Light Cut 225.9-244.5 °C	0.0046296	1.9239
Arabian Light Cut 244.5-262.5 °C	0.0050003	1.9437
Arabian Light Cut 262.5-278.7 °C	0.0051306	1.9016
Arabian Light Cut 278.7-294.5 °C	0.0055027	1.9159

(a) Φ values were obtained from Ref 1.

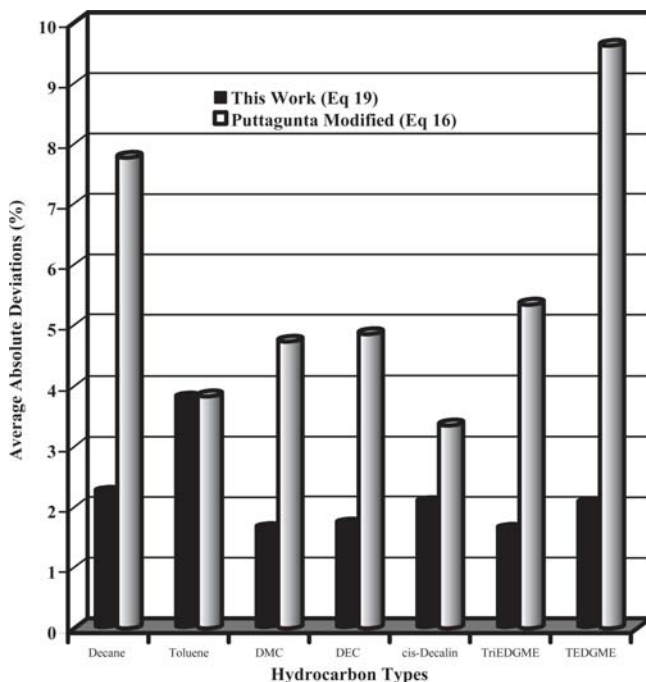


Fig. 1 Comparison of AADs between Eq 16 and 19 for various solvents

substituted with the constants Φ (as proposed by Miadonye and D’Orsay in Ref 1), Ψ , and Φ as shown in Eq 19:

$$\ln \eta = 2.3026 \left[\frac{b}{1 + \left(\frac{T - 37.78 \text{ }^\circ\text{C}}{310.93 \text{ }^\circ\text{C}} \right)^\Phi + C} \right] + \Psi P \exp(\Omega T) \quad (\text{Eq 19})$$

where

$$\Omega = -0.0033170 \quad (\text{Eq 20})$$

Ψ and Φ are constants that are unique to each sample and are referred to as the “pressure compressibility factor” on molecules and the “viscosity reduction factor,” respectively. This modification improves the predictability of this model for the previously mentioned hydrocarbons. The values of Ψ and Φ for these hydrocarbons are listed in Table 1.

The purpose of this paper is to present the results of the models, represented by Eq 16 and 19, tested on 15 hydrocarbon solvents, at temperatures ranging from -39.15 to 120 °C and at pressures ranging from 0.1 to 240 MPa. The reference temperature of 37.78 °C (100 °F) and the reference pressure of 1 atm as stipulated in Puttagunta’s model (Ref 2) were retained in Eq 16 and 19 due to practicality and expediency. It is convenient to use this temperature and this pressure as reference values when calculating kinematic viscosity because it requires minimal cost and effort to make measurements at these settings, and ASTM D45 standards (Ref 18) for field viscosity measurement are satisfied.

Values for the pressure parameters in Eq 19 were obtained by performing a constraint nonlinear regression on data collected from several literature sources shown in Table 2. The literature data consisted of 503 individual data points measured at varying pressures from 0.1 to 240 MPa.

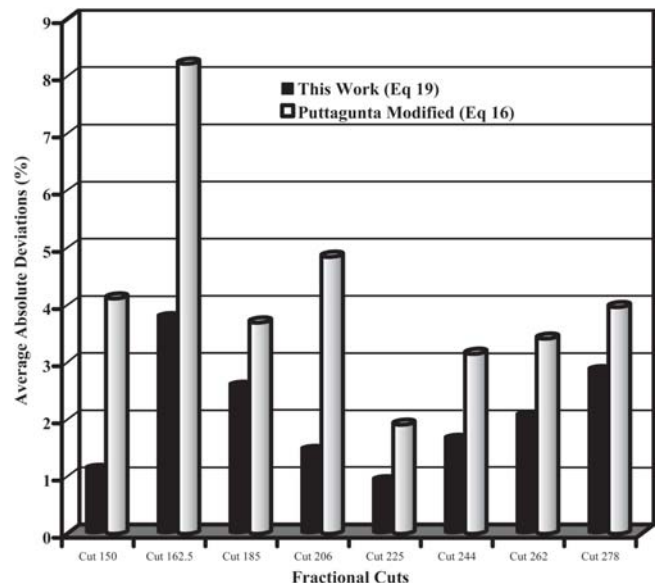


Fig. 2 Comparison of AADs between Eq 16 and 19 for Arabian light crude oil cuts referenced by their initial fractional distillation temperature

3. Results and Discussion

The two correlations discussed in this paper were evaluated with 503 data points obtained from 15 light hydrocarbon solvents. Solvent data temperatures range from -39.15 to 120 °C, and pressures range from atmospheric pressure up to 240 MPa. The AADs produced by the models represented by Eq 16 and 19 are 4.77 and 2.31%, respectively. Figures 1 and 2 illustrate the AADs obtained with both equations. A comparison of the accuracy of another model with the two equations is shown in Table 3.

Although the errors provided with Eq 1 are comparatively better than those obtained with Eq 16 and 19, it requires that a large number of parameters be determined. In Eq 16, the authors have maintained the simplicity of the Puttagunta et al. correlation (Ref 2) but derived new pressure parameters for light crude petroleum in a similar manner as described in an-

other of their publications (Ref 3). The errors obtained were very high and were characteristically higher as the pressure was increased. The viscosity-pressure relationship indicates an increase in viscosity with pressure at any given temperature without convergence. Thus, the pressure compressibility factor introduced in Eq 19 accounts for the pressure effects on viscosity. As illustrated in Fig. 3, 4, and 5, the correlation represented by Eq 19 produces reasonably low AADs between experimental and predicted viscosities based on the effect of temperature and pressure.

The AADs obtained with Eq 19, compared with Eq 16, are well within the experimental uncertainty of the data (Table 3 and 4). The results shown in Table 5 and 6, indicate that Eq 19 prediction accuracy improves for high temperature data. This arguably could be attributed to the increasing sensitivity of the pressure compressibility factor as the temperature increases.

These results show that this correlation (Eq 19) is very

Table 2 List of data sources

Hydrocarbons	No. of data points	Viscosity at 37.78 °C	Source
TriEGDME	32	1.6013	Comuñas et al. (Ref 4)
TEGDME	32	2.55999	Comuñas et al. (Ref 4)
Dimethyl carbonate	28	0.47273	Comuñas et al. (Ref 4)
Diethyl carbonate	32	0.66738	Comuñas et al. (Ref 4)
<i>cis</i> -Decalin	42	2.63076	Zéberg-Mikkelsen et al. (Ref 6)
Decane	40	0.98169	Audonnet and Pádua (Ref 8)
Toluene	75	0.59854	Assael et al. (Ref 9)
Toluene	96	0.58815	Assael et al. (Ref 10)
Toluene	18	0.59854	Caudwell et al. (Ref 19)
Arabian Light Cut 150-162.5 °C	9	0.79008	Kanti et al. (Ref 20)
Arabian Light Cut 162.5-185 °C	15	0.90942	Kanti et al. (Ref 20)
Arabian Light Cut 185-206.1 °C	12	1.19936	Kanti et al. (Ref 20)
Arabian Light Cut 206.1-225.9 °C	15	1.45942	Kanti et al. (Ref 20)
Arabian Light Cut 225.9-244.5 °C	12	1.82757	Kanti et al. (Ref 20)
Arabian Light Cut 244.5-262.5 °C	15	2.28615	Kanti et al. (Ref 20)
Arabian Light Cut 262.5-278.7 °C	15	2.79191	Kanti et al. (Ref 20)
Arabian Light Cut 278.7-294.5 °C	15	3.48391	Kanti et al. (Ref 20)

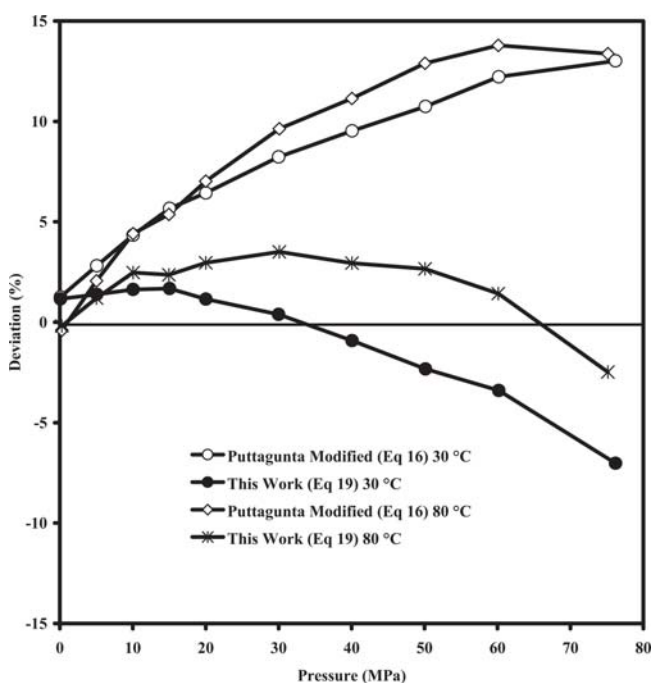


Fig. 3 Deviations of predicted viscosity data for decane

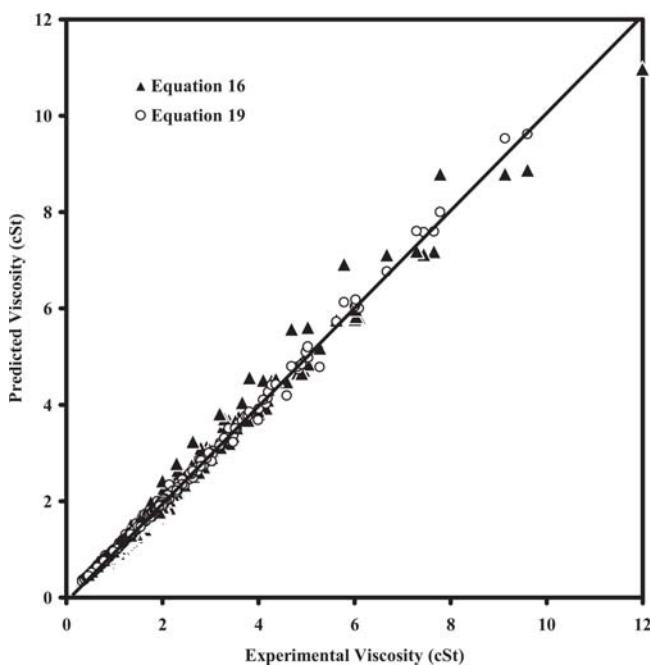


Fig. 4 Deviations of predicted viscosity calculated by Eq 16 and 19 from experimental viscosity

accurate in its ability to predict temperature and pressure effects on the kinematic viscosity of light hydrocarbon solvents. In addition, this model is composed of two constants that are unique to each hydrocarbon and one parameter that is deduced from a reference measurement, where as the other models re-

quire determination of five to nine parameters and/or the knowledge of four to eight physical property values.

4. Conclusions

The model the authors have presented in this paper, represented by Eq 19, is highly accurate in predicting the viscosity-temperature-pressure relationship of light hydrocarbon solvents. The results obtained from evaluating 503 data points indicate that the model predicts solvent viscosities within the limit of experimental uncertainty. With incorporation of a pressure compressibility factor, the model (Eq 19) gave an overall AAD of 2.31%, while an overall AAD of 4.77% was obtained (Eq 16) without it. Another advantage to this correlation is its versatility. The model only requires the calculation of two constants (the pressure constant and the shape factor constant) and one parameter that entails a reference measurement. This correlation contains fewer constants that need to be determined.

Table 3 Comparison of average absolute deviations (%) for several solvents

Sample	No. of data points	Average absolute deviation, %		
		Comuñas et al., Eq 1	Eq 16	Eq 19
Dimethyl carbonate	32	0.60	4.59	1.67
Diethyl carbonate	28	1.00	4.86	1.71
TEGDME	32	1.30	9.62	2.08
TriEGDME	32	0.90	4.97	1.65
Toluene	96	...	3.35	4.08

Table 4 Experimental viscosity and absolute percent deviation (%) calculated by Eq 16 and 19 at various pressures for decane at 50 °C

Pressure, MPa	Experimental viscosity, cSt	Deviation, %	
		Eq 16(a)	Eq 19(b)
0.38	0.8651	0.82	0.82
5.07	0.9093	2.62	1.46
10.06	0.9529	3.90	1.52
15.05	0.999	5.20	1.62
20.03	1.050	6.69	1.95
29.96	1.149	8.88	1.81
39.97	1.251	10.43	1.01
49.64	1.350	11.43	-0.30
59.43	1.455	12.22	-1.90
75.58	1.635	12.94	-5.27

(a) AAD = 7.51. (b) AAD = 1.77

Table 5 Experimental viscosity and absolute percent deviation (%) calculated by Eq 16 and 19 at various pressures for dimethyl carbonate at 50, 60 and 70 °C

Temperature, °C	Pressure, MPa	Experimental viscosity, cSt	Deviation, %	
			Eq 16(a)	Eq 19(b)
50	0.1	0.4323	-0.72	2.29
	20	0.4904	2.91	3.08
	40	0.5432	4.09	1.47
60	0.1	0.6000	5.02	-0.43
	20	0.3987	-2.70	2.42
	40	0.4432	-0.58	1.60
70	0.1	0.4968	2.28	1.55
	20	0.5452	3.01	-0.63
	40	0.3643	-6.22	0.83
70	0.1	0.4095	-2.44	1.47
	20	0.4544	-0.12	0.78
	40	0.5036	2.03	-0.04

(a) AAD = 2.68. (b) AAD = 1.38

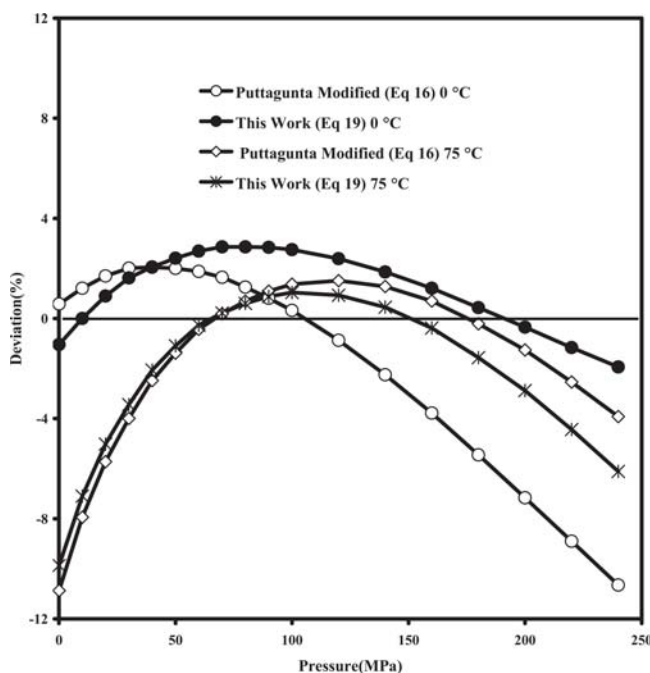


Fig. 5 Deviations of predicted viscosity data for toluene

Table 6 Predicted viscosity and absolute percent deviation (%) calculated by Eq 16 and 19 at various pressures for Arabian light crude oil cut 206.1-225.9 at 25, 40, 60, 80, and 100 °C

Temperature, °C	Pressure, MPa	Experimental viscosity, cSt	Deviation, %	
			Eq 16(a)	Eq 19(b)
25	0.1	1.815	1.85	1.64
	20	2.326	9.17	3.97
	40	2.775	9.64	-0.84
40	0.1	1.429	0.98	0.99
	20	1.786	6.60	1.97
	40	2.136	7.80	-1.59
60	0.1	1.099	0.96	1.17
	20	1.354	5.81	2.00
	40	1.602	6.72	-1.22
80	0.1	0.8467	-3.25	-2.87
	20	1.051	3.23	0.07
	40	1.256	5.82	-0.83
100	0.1	0.7065	-2.70	-2.20
	20	0.8575	2.24	-0.30
	40	1.0300	5.85	0.41

(a) AAD = 4.84. (b) AAD = 1.47

This greatly simplifies the calculations necessary to predict viscosity and provides enormous savings on computation time and cost.

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