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Viscosity Models Based on the Free Volume and Entropy Scaling Theories for Pure Hydrocarbons over a Wide Range of Temperatures and Pressures

Hseen O. Baled and Isaac K. Gamwo

Abstract

Viscosity is a critical fundamental property required in many applications in the chemical and oil industry. Direct measurements of this property are usually expensive and time-consuming. Therefore, reliable predictive methods are often employed to obtain the viscosity. In this work, two viscosity models based on the free-volume and entropy scaling theories are assessed and compared for pure hydrocarbons. The modeling results are compared to experimental data of 52 pure hydrocarbons including straight-chain alkanes, branched alkanes, cycloalkanes, and aromatics. This study considers viscosity data to extremely high-temperature and high-pressure (HTHP) conditions up to 573 K and 300 MPa. The results obtained with the free-volume theory viscosity in conjunction with the perturbed-chain statistical associating fluid theory (PC-SAFT) equation of state are characterized by an overall average absolute deviation (AAD%) of 3% from the experimental data. The overall AAD% obtained with the predictive entropy scaling method by Lötgering-Lin and Gross is 8%.

Keywords: high temperature, high pressure, hydrocarbons, modeling, viscosity

1. Introduction

Viscosity is a key property in many engineering disciplines, including chemical and petroleum engineering. For instance, viscosity influences the fluid flow through porous media and pipelines; hence, it is required for the design of pipelines and transport equipment as well as for the estimation of recoverable oil and flow rates in porous media or wellbores. Viscosity can be determined through experimental measurements. However, carrying out viscosity measurements at all conditions of interest is not only expensive and time-consuming but also may not be possible at extreme conditions such as those encountered in ultra-deep reservoirs including pressures up to 300 MPa and temperatures up to 573 K. Reliable prediction models provide an alternative approach to generating predicted and correlated viscosity data at conditions where experimental data are not readily available.

Unlike the viscosity of gases at low pressures which is well defined by the kinetic theory of the gases, the viscosity theory of liquids is still inadequately developed due to the complications caused by the intermolecular forces between the molecules [1]. Therefore, there is no widely accepted simple theoretical method for predicting liquid viscosities, and most estimation techniques used for viscosity prediction of liquids are of empirical or semiempirical nature. The empirical models are correlations based on experimental observation with no theoretical background, whereas semi-theoretical models have a fundamental basis but contain adjustable parameters determined by fitting the model to experimental data.

In the present study, two viscosity models based on the free-volume and entropy scaling theories are assessed and tested against viscosity data for pure hydrocarbons from different chemical families that are commonly found in crude oil at high-temperature and high-pressure (HTHP) conditions up to 573 K and 300 MPa. Pure components are well-suited for initial evaluation of the viscosity models because a viscosity model that has difficulty in correctly describing the viscosity of a single hydrocarbon is likely to fail when predicting multicomponent mixtures.

2. Free-volume theory (FVT)

The FVT model is based on the free-volume concept. The idea that the viscosity depends upon the free space was first introduced by Batschinski [2] about 100 years ago. The viscosity, η , can be expressed as a sum of two contributions given in Eq. (1):

$$\eta = \eta_0 + \Delta\eta \quad (1)$$

where η_0 is the dilute gas viscosity and the $\Delta\eta$ term dominates for liquid viscosity. The dilute gas term η_0 is determined from the kinetic gas theory at very low pressures. It should be noted, though, that for liquids and supercritical fluids, the dilute gas viscosity term η_0 is negligibly small in comparison to the total viscosity η ; hence, η_0 can be neglected for such fluids. Doolittle [3] found that the viscosity of liquid n-alkanes can be represented by a simple function of the free space fraction, $f_v = \frac{v_f}{v_0} = \frac{v-v_0}{v_0}$:

$$\eta = A \exp(B/f_v) \quad (2)$$

where v_0 is the molecular volume of reference or hard-core volume, v is the specific molecular volume, B is characteristic of the free-volume overlap, and A is a material-specific constant. A viscosity model based on the relation between free volume, friction coefficient, and viscosity has been proposed by Allal and coauthors [4, 5]:

$$\Delta\eta = A \exp(B/f_v) \quad (3)$$

where the free-volume fraction f_v was defined by means of the fluctuation-dissipation theory as

$$f_v \propto \left(\frac{RT}{E}\right)^{\frac{3}{2}} \quad (4)$$

where R is the gas constant and T is the temperature. In this expression, $E = E_0 + \frac{PM}{\rho}$, where $E_0 = \alpha\rho$ is related to the energy barrier that the molecule has to

overcome in order to diffuse, ρ is the density, and P is the pressure. The viscosity of the dense state is linked to the fluid microstructure using the friction coefficient, ζ , which is related to molecular mobility and to the diffusion of linear momentum:

$$\Delta\eta = \frac{\rho N_A \zeta L^2}{M} \quad (5)$$

where N_A is Avogadro's number and L is a characteristic molecular length parameter. By combining the relations between free volume, friction coefficient, and viscosity, the following expression was obtained for the viscosity:

$$\Delta\eta = \frac{\rho l \left(\alpha \rho + \frac{PM}{\rho} \right)}{\sqrt{3RTM}} \exp \left[B \left(\frac{\alpha \rho + \frac{PM}{\rho}}{RT} \right)^{3/2} \right] \quad (6)$$

The unit for the viscosity is [Pa·s], when all other variables are in SI units. The term $\frac{PM}{\rho}$ is linked to the energy necessary to form vacant vacuums required for the diffusion of the molecules. l is the characteristic length parameter in Å. The unitless parameter B is characteristic of the free-volume overlap. The density appears explicitly in Eq. (6), and hence the values of the free-volume theory parameters are directly dependent on whether experimental or calculated densities are used.

The three pure component parameters l , α , and B are determined by fitting Eq. (1) to experimental viscosity data. The use of the FVT requires density information, either experimental or calculated values. In this study, FVT is used in conjunction with the hybrid group-contribution perturbed-chain statistical associating fluid theory equation of state (G-C PC-SAFT EoS) [6] since this EoS provides reliable density predictions over wide ranges of pressure and temperature. In this equation, the PC-SAFT parameters are determined using two different sets of group-contribution (G-C) parameters for two pressure ranges: low-to-moderate pressures ($\lesssim 7$ MPa) and high pressures ($\gtrsim 7$ MPa).

3. Entropy scaling model by Lötgering-Lin and Gross (ES-LG)

The basic idea of this method is to relate the viscosity to the residual entropy. The residual entropy is defined as the difference between a real state value and ideal gas state value at the same temperature and density, $s_{res}(\rho, T) = s(\rho, T) - s^{id}(\rho, T)$. Lötgering-Lin and Gross [7] proposed a predictive entropy scaling method for viscosities using a group-contribution (G-C) method based on the group-contribution perturbed-chain polar statistical associating fluid theory equation of state (G-C PCP-SAFT EoS) [8, 9]. Lötgering-Lin and Gross linked the Chapman-Enskog viscosity to PCP-SAFT segments in terms of G-C parameters, with

$$\eta_{CE, gc} = \frac{5}{16} \frac{\sqrt{M_{mol} k_B T / (m_{gc} N_A \pi)}}{\sigma_{gc}^2 \Omega_{gc}^{(2,2)*}} \quad (7)$$

where M_{mol} is the molar mass, k_B is the Boltzmann constant, and T is the absolute temperature, N_A is Avogadro's number, m is the segment number, σ is the segment diameter, and $\Omega^{(2,2)*}$ is the reduced collision integral. The index gc indicates pure component parameters that are calculated with the group-contribution method based on G-C-PCP-SAFT EoS.

A reduced viscosity is then defined as

$$\eta^* = \frac{\eta}{\eta_{CE,gc}} \quad (8)$$

where the pure component reduced viscosity, η_i^* , is empirically correlated as

$$\ln \eta_i^* = A_i + B_i z + C_i z^2 + D_i z^3 \quad (9)$$

with

$$z = \frac{s_{res}}{k_B m_{gc,i}} \quad (10)$$

The residual entropy, s_{res} , is calculated from the G-C-PCP-SAFT EoS originally proposed by Vijande et al. [8] and reparametrized by Sauer et al. [9]:

$$s_{res}(\rho, T) = - \left(\frac{\partial a_{res}}{\partial T} \right)_\rho \quad (11)$$

where $a_{res} = A_{res}/N$ is the specific Helmholtz energy given by Gross and Sadowski [10]. N is the total number of molecules.

The viscosity parameters A_i to D_i of pure substances are obtained from parameters A_α to D_α of functional group α , respectively. The following empirical expressions are proposed by Lötgering-Lin and Gross [7] for mixing group-contribution parameters:

$$A_i = \sum_\alpha n_{\alpha,i} m_\alpha \sigma_\alpha^3 A_\alpha \quad (12)$$

$$B_i = \sum_\alpha \frac{n_{\alpha,i} m_\alpha \sigma_\alpha^3}{V_{tot,i}^\gamma} B_\alpha \quad (13)$$

$$C_i = \sum_\alpha n_{\alpha,i} C_\alpha \quad (14)$$

$$D_i = D \sum_\alpha n_{\alpha,i} \quad (15)$$

With

$$V_{tot,i} = \sum_\alpha n_{\alpha,i} m_\alpha \sigma_\alpha^3 \quad (16)$$

where $n_{\alpha,i}$ denotes the number of functional groups of type α in the substance i . The exponent γ and the parameter D are kept constant for all studied substances and are optimized for n-alkanes ($D = -0.01245$ and $\gamma = 0.45$) [7]. The group-contribution parameters A_α , B_α , and C_α of all groups α are given in [7].

4. Modeling results

The two viscosity methods, FVT and ES-LG, are tested on a database consisting of 52 hydrocarbons (21 normal alkanes, 13 branched alkanes, 4 cycloalkanes, 14 aromatics) typically present in most of the crude oils from ambient conditions to

extremely high-temperature and high-pressure (HTHP) conditions up to 573 K and 300 MPa. The temperature and pressure ranges considered in this study are given for each substance in **Table 1**. The performance of each model is assessed by the following statistical measures:

$$\text{Absolute Average Deviation (AAD)} = \frac{100}{N} \sum_{i=1}^N \left| \frac{\eta_{i, \text{cal}} - \eta_{i, \text{exp}}}{\eta_{i, \text{exp}}} \right| \quad (17)$$

$$\text{Maximum Deviation (MD)} = 100 \cdot \max \left| \frac{\eta_{i, \text{cal}} - \eta_{i, \text{exp}}}{\eta_{i, \text{exp}}} \right| \quad (18)$$

$$\text{Bias} = \frac{100}{N} \sum_{i=1}^N \frac{\eta_{i, \text{cal}} - \eta_{i, \text{exp}}}{\eta_{i, \text{exp}}} \quad (19)$$

where N is the total number of data points, $\eta_{i, \text{cal}}$ represents the calculated viscosity value, and $\eta_{i, \text{exp}}$ is the experimental data point obtained from the literature. The absolute average deviation AAD is a measure of how close the calculated values are to the experimental data, while the bias indicates how well the calculated values are distributed around the literature data. Low values of the bias imply that the deviations are evenly distributed about zero. A positive bias indicates overestimation of the calculated viscosity, whereas a negative value indicates

Compound	Ranges of conditions		Reference	FVT			ES-LG		
	T/K	P/MPa		AAD/%	MD/%	Bias/%	AAD/%	MD/%	Bias/%
Straight-chain alkanes (normal alkanes)									
CH ₄	298–573	0.1–300	[11–13]	3	33	–2	15	47	13
C ₂ H ₆	298–573	0.1–70	[11, 14]	2	5	1	7	35	–6
C ₃ H ₈	298–500	0.1–100	[14, 15]	5	21	1	6	20	–2
n-C ₄ H ₁₀	298–573	0.1–69	[11]	4	20	2	5	21	2
n-C ₅ H ₁₂	298–573	0.1–252	[16, 17]	3	18	1	6	15	–3
n-C ₆ H ₁₄	298–573	0.1–300	[16, 18]	4	18	–2	5	19	–1
n-C ₇ H ₁₆	298–573	0.1–100	[16, 19]	2	6	0	4	17	4
n-C ₈ H ₁₈	298–523	0.1–242	[18, 20, 21]	2	6	0	2	7	2
n-C ₉ H ₂₀	298–473	0.1–300	[16, 19, 21]	2	9	0	4	10	4
n-C ₁₀ H ₂₂	298–573	0.1–300	[16, 17, 21–24]	2	6	0	4	14	2
n-C ₁₁ H ₂₄	303–323	0.1–62	[19]	0.1	1	0	1	4	–1
n-C ₁₂ H ₂₆	298–573	0.1–300	[16, 22–27]	3	13	0	4	17	2
n-C ₁₃ H ₂₈	303–353	0.1–100	[28]	1	4	1	4	8	–4
n-C ₁₄ H ₃₀	313–393	0.69–60	[29]	2	8	–1	5	12	–2
n-C ₁₅ H ₃₂	310–408	0.1–320	[25]	2	7	0	4	11	0
n-C ₁₆ H ₃₄	298–534	0.1–273	[30, 31]	3	9	0	10	24	1
n-C ₁₇ H ₃₆	323–573	0.1–0.1	[3]	2	6	–2	8	16	–8
n-C ₁₈ H ₃₈	326–534	0.1–280	[25, 31]	3	12	0	9	28	8
n-C ₁₉ H ₄₀	333–523		[32]	3	8	0	4	10	2
n-C ₂₀ H ₄₂	326–534	1.38–243	[31, 33]	4	13	0	12	40	9

Compound	Ranges of conditions		Reference	FVT			ES-LG		
	T/K	P/MPa		AAD/%	MD/%	Bias/%	AAD/%	MD/%	Bias/%
n-C ₃₂ H ₆₆	373–458	0.3–0.3	[34]	1	2	0	2	4	2
Branched alkanes									
Isobutane	300–511	0.1–55	[35, 36]	3	11	2	3	13	–1
Isopentane	303–573	0.098–196	[16, 37]	6	22	–3	11	23	–7
Neopentane	311–444	0.7–55	[38]	7	23	–3	16	60	–13
2-Methylpentane	298–550	0.1–300	[16]	1	9	0	5	16	1
3-Methylpentane	313	0.1–147	[39]	1	4	1	8	10	8
2,2-Dimethylbutane	313	0.1–147	[39]	2	8	–1	27	37	–27
2,3-Dimethylbutane	313	0.1–147	[39]	0.3	1	0	7	11	–7
3-Ethylpentane	313	0.1–147	[39]	2	3	–2	17	20	17
2,4-Dimethylpentane	313	0.1–147	[39]	2	6	1	23	29	23
2,2,4-Trimethylpentane	298–523	0.1–300	[20, 40–42]	2	8	0	5	13	1
2,3,4-Trimethylpentane	298–453	0.1–195	[41]	3	11	0	7	15	–7
Squalane	303–473	1–202	[43, 44]	6	33	–2	26	49	23
2,2,4,4,6,8,8-Heptamethylnonane	298–453	0.1–195	[41, 45, 46]	7	49	–6	7	51	–5
Cycloalkanes									
Cyclopentane	298–353	0.1–300	[47, 48]	5	16	–2	2	6	–1
Cyclohexane	298–393	0.1–100	[49, 50]	2	7	0	4	14	1
Methylcyclohexane	298–343	0.1–300	[51, 52]	1	6	0	4	10	–4
Ethylcyclohexane	300–530	1–50	[53]	3	12	–1	7	23	–5
Aromatics									
Benzene	298–373	0.1–300	[54]	1	3	0	5	13	–4
Toluene	298–373	0.1–299	[55]	2	6	0	3	9	2
Ethylbenzene	298–453	0.1–195	[41]	2	4	1	16	40	16
Butylbenzene	313–373	0.1–100	[56]	2	9	0	4	11	4
Hexylbenzene	313–373	0.1–100	[56]	1	3	0	5	9	5
Octylbenzene	313–373	0.1–100	[56]	1	5	0	4	9	3
1,2-Diphenylethane	353–453	0.1–195	[41]	1	2	0	12	17	–12
m-Xylene	298–473	0.1–199	[20]	2	6	0	4	10	–2
o-Xylene	298–348	0.1–110	[57]	1	3	0	25	30	–25
p-Xylene	298–348	0.1–110	[57]	2	7	–1	6	8	–6
Naphthalene	375–454	0.1–101	[58]	8	33	1	11	22	–11
1-Methylnaphthalene	298–473	0.1–200	[20]	11	35	–7	32	60	–32
Tetralin	298–448	0.1–201	[20]	5	20	–1	29	53	28
Phenanthrene	396–573	0.1–101	[58]	13	41	–6	17	29	–17
Overall AAD%				3			8		

Table 1. Performance of FVT and ES-LG models for pure hydrocarbons over wide ranges of pressure and temperature (entries are rounded to nearest whole number).

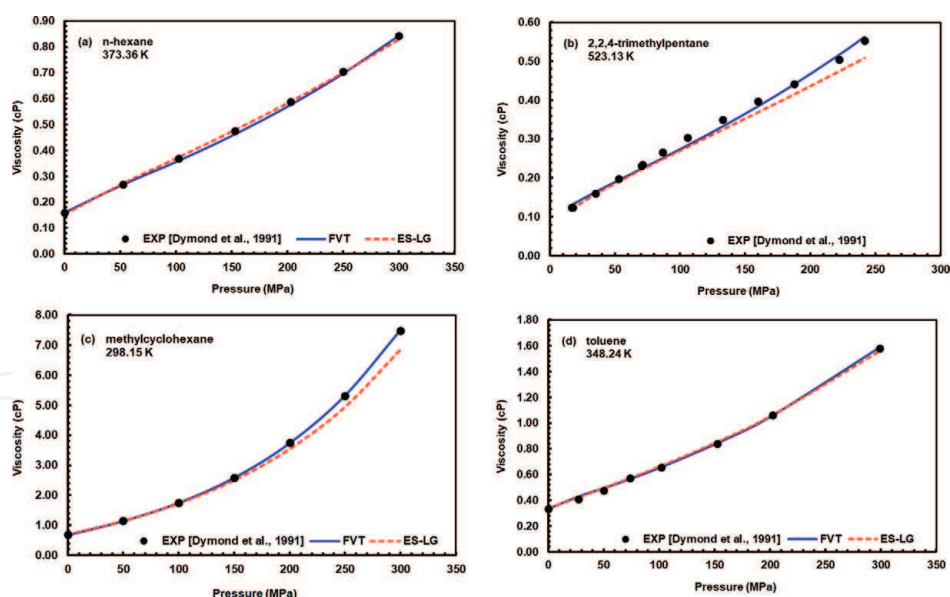


Figure 1. Viscosity predictions obtained with FVT and ES-LG viscosity models compared with literature data (EXP) for (a) n-hexane, (b) 2,2,4-trimethylpentane, (c) methylcyclohexane, and (d) toluene.

underestimation. These statistical measures of the ability of each of the selected seven models to reproduce viscosity values at HTHP conditions for each of the 52 pure compounds are given in **Table 1**.

The overall AAD obtained with the FVT model in conjunction with the hybrid G-C PC-SAFT EoS is 3%. The three adjustable parameters (l , α , B) required in this method are obtained by fitting the FVT predictions for each pure compound to the corresponding literature data. These optimized parameters yield reliable viscosity values over the whole ranges of temperatures to 573 K and pressures to 300 MPa.

The results obtained with the entropy scaling method by Lötgering-Lin and Gross are generally in good agreement with experimental data with an overall AAD of 8%. The AADs obtained for n-alkanes, branched alkanes, cycloalkanes, and aromatics are within 1–15, 3–27, 2–7, and 3–32%, respectively. These results are impressive for a fully predictive model that requires only the input of the molecular mass and the number of functional groups in each molecule. Unfortunately, this model cannot differentiate between isomers, such as 2-methylpentane and 3-methylpentane, and xylene isomers. In addition, this model has not yet been extended to binary, ternary, and multicomponent mixtures, such as crude oils.

For comparison purposes, **Figure 1(a–d)** shows the performance of the two studied viscosity methods, FVT and ES-LG, for four pure compounds representative of straight-chain alkanes (n-hexane), branched alkanes (2,2,4-trimethylpentane), cycloalkanes (methylcyclohexane), and aromatics (toluene).

5. Conclusions

This work provides an assessment of the capabilities of two viscosity methods based on the free-volume and entropy scaling theories to model the viscosity of pure hydrocarbons over wide ranges of temperatures and pressures. The performance of the two studied viscosity models is discussed and evaluated by comparison to experimental viscosity data of 52 pure hydrocarbons from four different chemical families, namely, straight-chain alkanes, branched alkanes, cycloalkanes, and aromatics, at ambient and extremely high-temperature and high-pressure (HTHP) conditions up to 573 K and 300 MPa. The viscosity of pure components is

required in most mixture models as an input parameter, and hence accurate and reliable model for pure compounds, particularly under high-pressure conditions, is a prerequisite for the mixture viscosity to be accurately estimated using the mixture model. The predictive entropy scaling method proposed by Lötgering-Lin and Gross (ES-LG model) predicts the viscosity with an overall absolute average deviation of about 8%, and the predictions are reasonable for most engineering and industrial applications given that the accuracy of most experimental viscosity data is within 1–5%. The free-volume theory (FVT) viscosity model provides very satisfactory results with an overall AAD of 3%. However, it is important to note that unlike the entropy scaling method, the free-volume theory is not a predictive model and requires that sufficient experimental viscosity data are readily available over the temperature and pressure ranges of interest to determine the fluid specific parameters.

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