We are IntechOpen, the world's leading publisher of Open Access books Built by scientists, for scientists

6,300

Open access books available

172,000

International authors and editors

190M

Downloads

Our authors are among the

154

Countries delivered to

TOP 1%

most cited scientists

12.2%

Contributors from top 500 universities

WEB OF SCIENCE™

Selection of our books indexed in the Book Citation Index
in Web of Science™ Core Collection (BKCI)

Interested in publishing with us?
Contact book.department@intechopen.com

Numbers displayed above are based on latest data collected.
For more information visit www.intechopen.com

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.

Acknowledgements

The Open Access Publishing fee for this chapter was fully paid by the University Library System, University of Pittsburgh, USA.

Author details

Hseen O. Baled^{1,2*} and Isaac K. Gamwo²

1 Department of Chemical and Petroleum Engineering, University of Pittsburgh, Pittsburgh, PA, USA

2 National Energy Technology Laboratory, Research and Innovation Center, U.S. Department of Energy, Pittsburgh, PA, USA

*Address all correspondence to: hob9@pitt.edu

IntechOpen

© 2019 The Author(s). Licensee IntechOpen. This chapter is distributed under the terms of the Creative Commons Attribution License (<http://creativecommons.org/licenses/by/3.0>), which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

References

- [1] Poling BE, Prausnitz JM, O'Connell JP. The Properties of Gases and Liquids. 5th ed. New York: McGraw-Hill; 2001
- [2] Batschinski AJ. Untersuchungen über die innere Reibung der Flüssigkeiten. Zeitschrift für Physikalische Chemie. 1913;**84**:644-650
- [3] Doolittle AK. Studies in Newtonian flow. II. The dependence of the viscosity of liquids on free-space. Journal of Applied Physics. 1951;**22**:1471-1475
- [4] Allal A, Moha-Ouchane M, Boned C. A new free volume model for dynamic viscosity and density of dense fluids versus pressure and temperature. Physics and Chemistry of Liquids. 2001;**39**:1-30
- [5] Allal A, Boned C, Baylaucq A. Free-volume viscosity model for fluids in the dense and gaseous states. Physical Review E. 2001;**64**:011203/1-011203/10
- [6] Burgess WA, Tapriyal D, Gamwo IK, Wu Y, McHugh MA, Enick RM. New group-contribution parameters for the calculation of PC-SAFT parameters for use at pressures to 276 MPa and temperatures to 533 K. Industrial and Engineering Chemistry Research. 2014;**53**:2520-2528
- [7] Lötgering-Lin O, Gross J. Group contribution method for viscosities based on entropy scaling using the perturbed-chain polar statistical associating fluid theory. Industrial and Engineering Chemistry Research. 2015;**54**:7942-7952
- [8] Vijande J, Pineiro MM, Bessieres D, Saint-Guirons H, Legido JL. Description of PVT behaviour of hydrofluoroethers using the PC-SAFT EOS. Physical Chemistry Chemical Physics. 2004;**6**: 766-770
- [9] Sauer E, Stavrou M, Gross J. Comparison between a homo- and a heterosegmented group contribution approach based on the perturbed-chain polar statistical associating fluid theory equation of state. Industrial and Engineering Chemistry Research. 2014;**53**:14854-14864
- [10] Gross J, Sadowski G. Perturbed-chain SAFT: An equation of state based on a perturbation theory for chain molecules. Industrial and Engineering Chemistry Research. 2001;**40**: 1244-1260
- [11] Younglove BA, Ely JF. Thermophysical properties of fluids. II. Methane, ethane, propane, isobutane, and normal butane. Journal of Physical and Chemical Reference Data. 1987;**16**: 577-798
- [12] Evers C, Lösch H, Wagner W. An absolute viscometer-densimeter and measurements of the viscosity of nitrogen, methane, helium, neon, argon, and krypton over a wide range of density and temperature. International Journal of Thermophysics. 2002;**23**: 1411-1439
- [13] van der Gulik PS, Mostert R, van den Berg HR. The viscosity of methane at 25°C up to 10 kbar. Physica A: Statistical Mechanics and its Applications. 1988;**151**:153-166
- [14] Seibt D, Vo K, Herrmann S, Vogel E, Hassel E. Simultaneous viscosity-density measurements on ethane and propane over a wide range of temperature and pressure including the near-critical region. Journal of Chemical & Engineering Data. 2011;**56**:1476-1493
- [15] Vogel E, Küchenmeister C, Bich E, Laesecke A. Reference correlation of the viscosity of propane. Journal of Physical

and Chemical Reference Data. 1998;27: 947-970

[16] Lemmon EW, McLinden MO, Friend DG. Thermophysical properties of fluid systems. In: Linstrom PJ, Mallard WG, editors. NIST Chemistry WebBook, NIST Standard Reference Database Number 69. Gaithersburg, MD: National Institute of Standards and Technology; retrieved 2018

[17] Oliveira C, Wakeham W. The viscosity of five liquid hydrocarbons at pressures up to 250 MPa. *International Journal of Thermophysics*. 1992;13: 773-790

[18] Dymond JH, Robertson J, Isdale JD. Transport properties of nonelectrolyte liquid mixtures-III. Viscosity coefficients for n-octane, n-decane, and equimolar mixtures of n-octane + n-dodecane and n-hexane + n-dodecane from 25 to 100°C at pressures up to the freezing pressure or 500 MPa. *International Journal of Thermophysics*. 1981;2:133-154

[19] Assael MJ, Papadaki M. Measurements of the viscosity of n-heptane, n-nonane, and n-undecane at pressures up to 70 MPa. *International Journal of Thermophysics*. 1991;12: 801-810

[20] Caudwell DR, Trusler JPM, Vesovic V, Wakeham WA. Viscosity and density of five hydrocarbon liquids at pressures up to 200 MPa and temperatures up to 473 K. *Journal of Chemical & Engineering Data*. 2009;54: 359-366

[21] Huber ML, Laesecke A, Xiang HW. Viscosity correlations for minor constituent fluids in natural gas: n-octane, n-nonane and n-decane. *Fluid Phase Equilibria*. 2005;228-229:401-408

[22] Knapstad B, Skjolsvik PA, Oye HA. Viscosity of pure hydrocarbons. *Journal*

of Chemical & Engineering Data. 1989; 34:37-43

[23] Knapstad B, Skjolsvik PA, Oye HA. Viscosity of three binary hydrocarbon mixtures. *Journal of Chemical & Engineering Data*. 1991;36:84-88

[24] Dymond JH, Young KJ. Transport properties of nonelectrolyte liquid mixtures. I. Viscosity coefficients for n-alkane mixtures at saturation pressure from 283 to 378 K. *International Journal of Thermophysics*. 1980;1:331-344

[25] Hogenboom DL, Webb W, Dixon JA. Viscosity of several liquid hydrocarbons as a function of temperature, pressure, and free volume. *The Journal of Chemical Physics*. 1967; 46:2586-2598

[26] Huber ML, Laesecke A, Perkins RA. Transport properties of n-dodecane. *Energy & Fuels*. 2004;18:968-975

[27] Giller EB, Drikamer HG. Viscosity of normal paraffins near the freezing point. *Industrial and Engineering Chemistry*. 1949;41:2067-2069

[28] Daugé P, Canet X, Baylaucq A, Boned C. Measurements of the density and viscosity of the tridecane + 2,2,4,4,6,8,8-heptamethylnonane mixtures in the temperature range 293.15-353.15 K at pressures up to 100 MPa. *High Temperatures - High Pressures*. 2001;33:213-230

[29] Hernández-Galván MA, García-Sánchez F, Macías-Salinas R. Liquid viscosities of benzene, n-tetradecane, and benzene + n-tetradecane from 313 to 393 K and pressures up to 60 MPa: Experiment and modeling. *Fluid Phase Equilibria*. 2007;262:51-60

[30] Dymond JH, Young KJ, Isdale JD. Transport properties of nonelectrolyte liquid mixtures-II. Viscosity coefficients for the n-hexane + n-hexadecane system at temperatures from 25 to 100°C at

pressures up to the freezing pressure or 500 MPa. *International Journal of Thermophysics*. 1980;1(4):345-373

[31] Baled HO, Xing D, Katz H, Tapriyal D, Gamwo IK, Soong Y, et al. Viscosity of n-hexadecane, n-octadecane and n-eicosane at pressures up to 243 MPa and temperatures up to 534 K. *The Journal of Chemical Thermodynamics*. 2014;72:108-116

[32] Guseinov S, Naziev Y. *Izvestiya Vysshikh Uchebnykh Zavedenii, Russian (USSR)*. Neft i Gaz. 1974;17:38-62

[33] Rodden JB, Erkey C, Akgerman A. High-temperature diffusion, viscosity, and density measurements in n-eicosane. *Journal of Chemical & Engineering Data*. 1988;33:344-347

[34] Aasen E, Rytter E, Oye HA. Viscosity of n-hydrocarbons and their mixtures. *Industrial and Engineering Chemistry Research*. 1990;29:1635-1640

[35] Diller DE, Van Poolen LJ. Measurements of the viscosities of saturated and compressed liquid normal butane and isobutane. *International Journal of Thermophysics*. 1985;6:43-62

[36] Gonzalez MH, Lee AL. Viscosity of isobutene. *Journal of Chemical & Engineering Data*. 1966;11:357-359

[37] Bridgman PW. The effect of pressure on the viscosity of forty-three pure liquids. *Proceedings of the American Academy of Arts and Sciences*. 1926;61:57-99

[38] Gonzalez MH, Lee AL. Viscosity of 2,2-dimethylpropane. *Journal of Chemical & Engineering Data*. 1968;13:66-69

[39] Kuss E, Pollmann P. Viskositäts-Druckabhängigkeit und Verzweigungsgrad flüssiger Alkane. *Zeitschrift für Physikalische Chemie*. 1969;68:205-227

[40] Dymond JH, Glen NF, Isdale JD. Transport properties of nonelectrolyte liquid mixtures-VII. Viscosity coefficients for isooctane and for equimolar mixtures of isooctane + n-octane and isooctane + n-dodecane from 25 to 100°C at pressures up to 500 MPa or to the freezing pressure. *International Journal of Thermophysics*. 1985;6:233-250

[41] Krahn UG, Luft G. Viscosity of several liquid hydrocarbons in the temperature range 298-453 K at pressures up to 200 MPa. *Journal of Chemical & Engineering Data*. 1994;39:670-672

[42] Pádua AAH, Fareleira JMNA, Calado JCG, Wakeham WA. Density and viscosity measurements of 2,2,4-trimethylpentane (isooctane) from 198 K to 348 K and up to 100 MPa. *Journal of Chemical & Engineering Data*. 1996;41:1488-1494

[43] Schmidt KAG, Pagnutti D, Curran MD, Singh A, Trusler JPM, Maitland GC, et al. New experimental data and reference models for the viscosity and density of squalane. *Journal of Chemical & Engineering Data*. 2015;60:137-150

[44] Ciotta F, Maitland G, Smietana M, Trusler JPM, Vesovic V. Viscosity and density of carbon dioxide + 2,6,10,15,19,23-hexamethyltetracosane (squalane). *Journal of Chemical & Engineering Data*. 2009;54:2436-2443

[45] Canet X, Daugé P, Baylaucq A, Boned C, Zéberg-Mikkelsen C, Quiñones-Cisneros SE, et al. Density and viscosity of the 1-methylnaphthalene + 2,2,4,4,6,8,8-heptamethylnonane system from 293.15 to 353.15 K at pressures up to 100 MPa. *International Journal of Thermophysics*. 2001;22:1669-1689

[46] Et-Tahir A, Boned C, Lagourette B, Xans P. Determination of the viscosity of various hydrocarbons and mixtures

of hydrocarbons versus temperature and pressure. *International Journal of Thermophysics*. 1995;**16**:1309-1334

[47] Harris KR, Newitt PJ, Woolf LA. Temperature and density dependence of the viscosity of cyclopentane. *Journal of Chemical & Engineering Data*. 2004;**49**: 138-142

[48] Kurihara K, Kandil ME, Marsh KN, Goodwin ARH. Measurement of the viscosity of liquid cyclopentane obtained with a vibrating wire viscometer at temperatures between (273 and 353) K and pressures below 45 MPa. *Journal of Chemical & Engineering Data*. 2007;**52**:803-807

[49] Hernández-Galván MA, García-Sánchez F, García-Flores BE, Castro-Arellano J. Liquid viscosities of cyclohexane, cyclohexane + tetradecane, and cyclohexane + benzene from (313 to 393) K and pressures up to 60 MPa. *Journal of Chemical & Engineering Data*. 2009;**54**:2831-2838

[50] Tanaka Y, Hosokawa H, Kubota H, Makita T. Viscosity and density of binary mixtures of cyclohexane with n-octane, n-dodecane, and n-hexadecane under high pressures. *International Journal of Thermophysics*. 1991;**12**: 245-264

[51] Jonas J, Hasha D, Huang SG. Self-diffusion and viscosity of methylcyclohexane in the dense liquid region. *The Journal of Chemical Physics*. 1979;**71**:3996-4000

[52] Baylaucq A, Boned C, Dauge P, Lagourette B. Measurements of the viscosity and density of three hydrocarbons and the three associated binary mixtures versus pressure and temperature. *International Journal of Thermophysics*. 1997;**18**:3-23

[53] Stephan K, Lucas K. Viscosity of Dense Fluids. 1st ed. New York: Plenum Press; 1979

[54] Dymond JH, Robertson J, Isdale JD. Transport properties of nonelectrolyte liquid mixtures - IV. Viscosity coefficients for benzene, perdeuterobenzene, hexafluorobenzene, and an equimolar mixture of benzene + hexafluorobenzene from 25 to 100°C at pressures up to the freezing pressure. *International Journal of Thermophysics*. 1981;**2**:223-236

[55] Dymond JH, Awan MA, Glen NF, Isdale JD. Transport properties of nonelectrolyte liquid mixtures. VIII. Viscosity coefficients for toluene and for three mixtures of toluene + hexane from 25 to 100°C at pressures up to 500 MPa. *International Journal of Thermophysics*. 1991;**12**:275-287

[56] Ducoulombier D, Zhou H, Boned C, Peyrelasse J, Saint-Guirons H, Xans P. Pressure (1-1000 bars) and temperature (20-100°C) dependence of the viscosity of liquid hydrocarbons. *The Journal of Physical Chemistry*. 1986;**90**:1692-1700

[57] Kashiwagi H, Makita T. Viscosity of twelve hydrocarbon liquids in the temperature range 298-348 K at pressures up to 110 MPa. *International Journal of Thermophysics*. 1982;**3**: 289-305

[58] Roetling JA, Gebhardt JJ, Rouse FG. Effect of pressure on viscosity of naphthalene, phenanthrene and impregnation pitches. *Carbon*. 1987;**25**: 233-242