



City Research Online

City St George's, University of London

Citation: Rokni, H. B., Moore, J. D. & Gavaises, E. (2021). Entropy-scaling based pseudo-component viscosity and thermal conductivity models for hydrocarbon mixtures and fuels containing iso-alkanes and two-ring saturates. *Fuel*, 283, 118877. doi: 10.1016/j.fuel.2020.118877

This is the accepted version of the paper.

This version of the publication may differ from the final published version. To cite this item please consult the publisher's version.

Permanent repository link: <https://openaccess.city.ac.uk/id/eprint/24686/>

Link to published version: <https://doi.org/10.1016/j.fuel.2020.118877>

Copyright and Reuse: Copyright and Moral Rights remain with the author(s) and/or copyright holders. Copies of full items can be used for personal research or study, educational, or not-for-profit purposes without prior permission or charge, unless otherwise indicated, provided that the authors, title and full bibliographic details are credited, a hyperlink and/or URL is given for the original metadata page and the content is not changed in any way. For full details of reuse please refer to [City Research Online policy](#).

1 **Entropy-scaling based pseudo-component viscosity and**
2 **thermal conductivity models for hydrocarbon mixtures and**
3 **fuels containing iso-alkanes and two-ring saturates**

4
5 *Houman B. Rokni^{1, 2}, Joshua D. Moore¹, and Manolis Gavaises²*

6
7 ¹ Afton Chemical Corp., Richmond, Virginia 23219, USA

8 ²Department of Mechanical Engineering and Aeronautics, City, University of London, Northampton
9 Square, EC1V 0HB London, UK

10
11
12 **Abstract**

13 Recently, Rokni et al. [1, 2] developed entropy-scaling based pseudo-component techniques to
14 predict the viscosity and thermal conductivity of hydrocarbon mixtures and fuels up to high
15 temperature and pressure conditions using only two calculated or measured mixture properties
16 (number average molecular weight and hydrogen-to-carbon ratio). The models are accurate for
17 many hydrocarbon mixtures that do not contain branched compounds (7 and 2% mean absolute
18 percent deviation (MAPD) for viscosity and thermal conductivity, respectively, on average).
19 However, predictions for some hydrocarbon mixtures and fuels containing iso-alkanes are often
20 less accurate (16 and 19% MAPD for viscosity and thermal conductivity, respectively, on average).
21 To improve predictions, it was proposed [1, 2] to fit one model parameter using an experimental
22 reference viscosity or thermal conductivity data point, which may not be ideal if experimental
23 reference data are not available. In order to make these models more practical, this study fits
24 empirical correlations for the model parameters, so that accurate predictions can be made without

¹Corresponding author, e-mail: joshua.moore@aftonchemical.com

25 fitting model parameters. The correlations enable viscosity and thermal conductivity predictions
26 for a wide range of hydrocarbon mixtures and fuels, including those containing branched alkanes,
27 and no longer require input of any experimental reference viscosity or thermal conductivity data.
28 The correlations are temperature (fit to data from 288 to 550 K) and pressure (fit to data from 1 to
29 4,400 bar) dependent and are functions of average molecular weight, hydrogen-to-carbon ratio, iso-
30 alkane and two-ring saturate concentrations. Viscosity and thermal conductivity predictions were
31 found to improve to within 5 and 2% average MAPD, respectively, relative to experimental data
32 for the hydrocarbon mixtures and fuels considered in this study.

33

34 **Introduction**

35 Thermophysical property models are used within computational fluid dynamics (CFD) to
36 assess the performance, emissions, and fuel economy of engines [3-10]. The accuracy of these CFD
37 models depends on accurate representation of the thermophysical fuel properties, especially
38 viscosity and thermal conductivity, often up to extreme pressure conditions. Various viscosity [1,
39 11, 12] and thermal conductivity [2, 13, 14] models have been proposed for fuels in the literature.
40 These models range from empirical [15-22] to mixture [23] and pseudo-component models [1, 2,
41 13, 14, 24-35], such as expanded fluid theory (EFT) [13, 14, 25-28], friction theory (FT) [29-33],
42 and free volume theory (FVT) [34, 35].

43 Recently, Rokni et al. [1, 2] developed an entropy-scaling based pseudo-component
44 (ESBPC) technique to predict viscosity and thermal conductivity of well-characterized
45 hydrocarbon mixtures and fuels (including rocket propellant, jet, and diesel fuels) up to high
46 temperature and high pressure (HTHP) conditions, using the perturbed-chain statistical associating
47 fluid theory (PC-SAFT) equation of state (EoS) [36]. The models define a single pseudo-component
48 to represent the compounds in a hydrocarbon mixture or fuel and at minimum, require the input of

49 two experimentally measured or calculated inputs (two-input parameter (2IP) model): the mixture
50 number average molecular weight (MW_{mixture}) and hydrogen to carbon ratio (HN/CN). While the
51 2IP model is accurate for many hydrocarbon mixtures containing components from different
52 chemical families (e.g., normal-alkanes, benzenes, naphthalenes, cyclohexanes), viscosity and
53 thermal conductivity predictions for some mixtures containing iso-alkanes are less accurate, with
54 up to 24% mean absolute percent deviation (MAPD) calculated for diesel, rocket propellant and jet
55 fuels [1, 2]. Predictions are improved for mixtures containing branched alkanes by fitting one model
56 parameter to a data point at a reference condition (e.g., data point at the lowest available temperature
57 and pressure). This three-input parameter (3IP) model predicted viscosity and thermal conductivity
58 with 4 and 2% MAPD on average, respectively, for several hydrocarbon mixtures and fuels [1, 2].

59 However, experimental viscosity and thermal conductivity reference data are often not
60 available for every mixture or fuel. The ESBPC technique would be more practical to use if the
61 accuracy of predictions were improved for hydrocarbon mixtures and fuels containing branched
62 alkanes but did not require fitting model parameters or input of experimental viscosity or thermal
63 conductivity reference data. This study achieves this by replacing the ESPBC model's thermal
64 conductivity and viscosity coefficients with empirical correlations, such that accurate viscosity and
65 thermal conductivity predictions up to HTHP conditions no longer require fitting of model
66 parameters or input of experimental viscosity or thermal conductivity reference data, even for
67 hydrocarbon mixtures and fuels containing branched alkanes. These empirical correlations
68 explicitly account for concentrations of iso-alkanes and two-ring saturates, such as decalin
69 derivative compounds. This Entropy Scaling Coefficient Corrected (ESCC) model requires three
70 mixture property inputs (MW_{mixture} , HN/CN, and weight fraction of iso-alkanes in the mixture
71 ($w_{\text{iso-alkanes}}$)) for thermal conductivity predictions and four mixture property inputs

72 (MW_{mixture} , HN/CN, $w_{\text{t iso-alkanes}}$, and weight fraction of two-ring saturates in the mixture
73 ($w_{\text{t two-ring saturates}}$)) for viscosity predictions. The iso-alkane and two-ring saturate chemical
74 families are selected based upon understanding that the ESPBC technique does not distinguish the
75 difference between normal and branched alkanes [1, 2]. Including the two-ring saturates in the
76 correlation is observed to improve the viscosity predictions. When using the ESCC model,
77 viscosity predictions (298 to 533 K and up to 2,600 bar) are within 5% MAPD, on average, which
78 is comparable to the uncertainty of the experimental data for the hydrocarbon mixtures (2%) and
79 diesel fuels (up to 4%) in this study. The thermal conductivity predictions (288 to 550 K and up to
80 4,400 bar) are 2% MAPD, on average, for the ESCC model, which is comparable to the uncertainty
81 of the experimental data for the hydrocarbon mixtures (1%) and rocket propellant and jet fuels (3%)
82 in this study.

83

84 **Hydrocarbon Mixtures**

85 Table 1 lists the studies which measured the viscosity of hydrocarbon mixtures up to high
86 pressures and temperatures. Dauge et al. [37] reported viscosity measurements for seven
87 compositions of binary mixtures (referred to as M1) containing 2,2,4,4,6,8,8-heptamethylnonane
88 (i.e., isocetane, referred to as HMN) and n-tridecane (nC13) from 293 to 353 K and up to 1,000 bar.
89 Zéberg-Mikkelsen et al. [38] measured viscosity of thirteen compositions of ternary mixtures
90 (referred to as M2) containing methyl-cyclohexane (MCH), decalin, and HMN from 293 to 353 K
91 and up to 1,000 bar. Ducoulombier et al. [39] reported viscosity for a quaternary mixture (referred
92 to as M3) containing n-decane (nC10), n-dodecane (nC12), n-tetradecane (nC14), and n-
93 hexadecane (nC16) from 313 to 353 K and up to 1,000 bar. Zéberg-Mikkelsen et al. [40] reported
94 viscosity measurements for 21 compositions of ternary mixtures (referred to as M4) containing

95 HMN, nC13, and methylnaphthalene (MNP) up to 353 K and 1,000 bar. Boned et al. [41] reported
 96 viscosity measurements for a ternary (referred to as M5) and a quinary (referred to as M6) mixture
 97 from 293 to 353 K and up to 1,000 bar.

98
 99 **Table 1.** Weight fractions of compounds in the mixtures considered in this study for viscosity
 100 [37-41].

Compound	Chemical Family	M1	M2	M3	M4	M5	M6
n-heptane	n-alkanes	-	-	-	-	-	-
n-decane	n-alkanes	-	-	0.193	-	-	-
n-dodecane	n-alkanes	-	-	0.231	-	-	-
n-tridecane	n-alkanes	0.104-0.851	-	-	0.109-0.750	0.400	0.200
n-tetradecane	n-alkanes	-	-	0.269	-	-	-
n-hexadecane	n-alkanes	-	-	0.307	-	-	-
2,2,4,4,6,8,8-heptamethylnonane	iso-alkanes	balance	balance	-	0.154-0.806	-	0.200
methylcyclohexane	cyclohexanes	-	0.062-0.618	-	-	-	-
heptylcyclohexane	cyclohexanes	-	-	-	-	0.349	0.350
decalin	decalins	-	0.087-0.719	-	-	-	-
heptyl benzene	benzenes	-	-	-	-	0.250	0.150
methylnaphthalene	naphthalenes	-	-	-	balance	-	0.100

101
 102 Table 2 lists the studies which report the thermal conductivity of hydrocarbon mixtures up
 103 to high temperatures and pressures. Wakeham et al. [42] reported thermal conductivity for binary
 104 mixtures containing benzene and trimethylpentane (TMP) (referred to as M7) for two different
 105 compositions at temperatures from 313 to 345 K and pressures up to 3,500 bar. Fareleira et al. [43]
 106 and Wakeham et al. [42] reported thermal conductivity data for binary mixtures containing n-
 107 heptane (C7) and TMP (referred to as M8) for three different compositions at temperatures from
 108 308 to 360 K and pressures up to 4,500 bar. Wada et al. [44] reported thermal conductivity data for
 109 binary mixtures containing C7 and n-undecane (C11), C7 and C16, and C11 and C16 (referred to
 110 as M9, M10, and M11, respectively) for three different compositions for each mixture at
 111 atmospheric pressure and temperatures from 295 to 345 K. Wada et al. [44] also reported thermal

112 conductivity for ternary mixtures (referred to as M12) including C7, C11, and C16 for three
 113 compositions at a range of temperatures from 295 to 345 K and 1 bar.

114
 115 **Table 2.** Weight fractions of compounds in the mixtures considered in this study for thermal
 116 conductivity [42-44].

Compound	Chemical Family	M7	M8	M9	M10	M11	M12
n-heptane	n-alkanes	-	0.456-0.726	0.658-0.342	-	0.430-0.871	0.052-0.462
n-undecane	n-alkanes	-	-	balance	0.187-0.674	-	0.242-0.361
n-hexadecane	n-alkanes	-	-	-	balance	balance	balance
trimethylpentane	iso-alkanes	0.328-0.814	balance	-	-	-	-
benzene	benzenes	balance	-	-	-	-	-

117
 118 **Fuels**
 119 Aquing et al. [11] reported viscosity of two diesel fuels (referred to as Middle East Straight
 120 Run (MESR) and Highly Naphthenic (HNA)) from 323 to 423 K and up to 3,500 bar. Rowane et
 121 al. [45] measured viscosity of three diesel fuels (referred to as Highly Paraffinic (HPF), Ultra-Low
 122 Sulfur Diesel (ULSD), and Highly Aromatic (HAR)) up to 533 K and 3,000 bar. Akhmedova-
 123 Azizova et al. [46] reported the thermal conductivity of RP1 fuel at temperatures between 293 and
 124 598 K and pressures up to 600 bar. Bruno [47] measured the thermal conductivity of RP2 fuel over
 125 a wide range of temperatures from 300 to 550 K and pressures up to 600 bar. Xu et al. [48] reported
 126 thermal conductivity measurements of RP3 fuel at temperatures from 285 to 513 K and pressures
 127 up to 50 bar. Jia et al. [49] reported the thermal conductivity of RP3 fuel at temperatures from 311
 128 to 399 K and a single isobar at 30 bar. Bruno [50] also measured the thermal conductivity of three
 129 different jet fuels, including JP-8 3773 (referred to as JP-8) and Jet A 4658 (referred to as Jet A) at
 130 high temperatures from 270 to 470 K and pressures up to 400 bar. The detailed chemical family
 131 compositions of the rocket propellant and jet fuels are reported elsewhere (RP1 [51], RP2 [51], RP3
 132 [51], Jet A [52], and JP-8 [52]).

133 **ESBPC Technique for Viscosity and Thermal Conductivity Predictions:**

134 The two-input parameter (2IP) techniques for viscosity and thermal conductivity predictions
135 are briefly described here but are explained in greater detail in ref. [1, 2]. Residual entropy, \tilde{s}^{res} ,
136 (the difference between the real fluid and ideal gas entropy) (Eq. (1)) (i.e., molar residual entropy,
137 \bar{s}^{res} , divided by the gas constant, R), in temperature (T) and volume (V) state variables, is calculated
138 from the residual Helmholtz free energy, \tilde{a}^{res} (the difference between the real fluid and ideal gas
139 Helmholtz free energy), using the PC-SAFT EoS.

140

$$\tilde{s}^{\text{res}}(V, T) = - \left(\frac{\partial \tilde{a}^{\text{res}}}{\partial T} \right)_V \quad (1)$$

141

142 The pseudo-component PC-SAFT parameters, m , σ , and ε/k , needed to calculate \tilde{s}^{res} are
143 determined using the pseudo-component technique and correlations shown in the supporting
144 information (SI). Using entropy-scaling, first proposed by Rosenfeld [53], a third-order polynomial,
145 proposed by Lötgering-Lin and Gross [54] (Eq. 2), is used to correlate the reduced transport
146 property (i.e., α^* , viscosity (η) or thermal conductivity (λ)) to reduced residual entropy (s^*) (Eq.
147 3).

148

$$\ln(\alpha^*) = \ln(\alpha/\alpha_{CE}) = A + Bs^* + Cs^{*2} + Ds^{*3} \quad (2)$$

$$s^* = \left(\frac{\tilde{s}^{\text{res}}(V, T)}{m} \right) \quad (3)$$

149

150 In Eq. (2), α_{CE} is the Chapman-Enskog viscosity or thermal conductivity [55-57]. The coefficients
151 (i.e., A , B , C , and D) in Eq. (2) are first calculated for an n-alkane and a poly-nuclear aromatic
152 (PNA) with the same number average molecular weight as the mixture (MW_{mixture}) using
153 correlations shown in the SI. Next, the pseudo-component coefficients are calculated by averaging

154 contributions of the n-alkane and PNA (using Eqs. (4 and 5) for viscosity and thermal conductivity
155 models, respectively), which uses an averaging parameter, Z , defined as a function of the mixture
156 degree of unsaturation ($\text{DoU}_{\text{mixture}}$). The procedure to calculate Z is provided in the SI.

157

$$\left(F \times m^2\right)_{\text{pseudo-component}} = (1 - Z) \times \left(F \times m^2\right)_{\text{n-alkane}} + Z \times \left(F \times m^2\right)_{\text{PNA}} \quad (4)$$

158

$$(F)_{\text{pseudo-component}} = (1 - Z) \times (F)_{\text{n-alkane}} + Z \times (F)_{\text{PNA}} \quad (5)$$

159
160 where F is the coefficient A , B , C , or D in Eq. (2). For mixtures containing iso-alkanes, the three-
161 input parameter (3IP-1) model, proposed in ref. [1, 2], fits one model parameter (D for viscosity or
162 B for thermal conductivity) to reproduce a single data point at a chosen reference state, instead of
163 calculating it using Eqs. (4 and 5). All model parameters needed to predict viscosity and thermal
164 conductivity for all hydrocarbon mixture compositions and fuels, in this study, are reported in the
165 SI.

166
167 **Entropy Scaling Coefficient Corrected (ESCC) Model**

168 In this study, a new model is proposed (referred to as the Entropy Scaling Coefficient
169 Corrected (ESCC) model), which correlates the pseudo-component coefficients, instead of
170 calculating them using Eqs. (4 and 5). The pseudo-component coefficients are correlated to mixture
171 properties ($\text{MW}_{\text{mixture}}$ and HN/CN) and weight fractions of iso-alkanes (i.e., heptamethylnonane
172 and trimethylpentane in this study) and two-ring saturates (i.e., decalin in this study). Training set
173 data was used to fit the G_i coefficients in Eq. (6) for viscosity and Eq. (7) for thermal conductivity.
174 The training set for the ESCC viscosity coefficient correlations (Eq. 6) contained a subset of
175 mixtures M1, M2, M4 as well as mixtures M3, M5, and M6 (690 total data points). The training set
176 for the ESCC thermal conductivity coefficient correlations (Eq. 7) contained a subset of mixtures

177 M7, M8, M9, M10, and M12 (134 total data points). The correlations were then validated for the
 178 remaining mixtures and fuels as a test set (356 thermal conductivity data points and 1134 viscosity
 179 data points). Tables 3 and 4 list the parameters in Eqs. (6 and 7) for the viscosity and thermal
 180 conductivity ESCC models, respectively.

$$G_{\eta} = G_{\eta_1} \times MW_{mixture} + G_{\eta_2} \times \frac{HN}{CN} + G_{\eta_3} \times wt_{iso-alkanes} + G_{\eta_4} \times wt_{two-ring\ saturates} + G_{\eta_5} \quad (6)$$

$$G_{\lambda} = G_{\lambda_1} \times MW_{mixture} + G_{\lambda_2} \times \frac{HN}{CN} + G_{\lambda_3} \times wt_{iso-alkanes} + G_{\lambda_4} \quad (7)$$

181
 182 In Eqs. (6 and 7) G_{η} and G_{λ} are the pseudo-component coefficients (A , B , C , or D) in Eq. (2), which
 183 are a function of $MW_{mixture}$, HN/CN , and weight fraction (wt) of iso-alkanes ($wt_{iso-alkanes}$) and
 184 two-ring saturates ($wt_{two-ring\ saturates}$). Inclusion of the two-ring saturate concentration was
 185 observed to improve the viscosity predictions using the ESCC correlation. For thermal conductivity
 186 predictions, a lack of available hydrocarbon mixture data containing two-ring saturates in the
 187 literature precluded including $wt_{two-ring\ saturates}$ in Eq. (7). The multilinear regression in Eqs. (6
 188 and 7) yielded satisfactory predictions for all mixture compositions based upon MAPD.

189
 190 **Table 3.** Parameters needed to calculate the coefficients (G_{η_i}) in Eq. (6) as a function of
 191 $MW_{mixture}$, HN/CN , $wt_{iso-alkanes}$, and $wt_{two-ring\ saturates}$ for the viscosity ESCC model: An
 192 average MAPD of 4.0% is obtained for viscosities using Eq. (6) for the mixture compositions
 193 used in the training set (690 data points).

G_{η_i}	G_{η_1}	G_{η_2}	G_{η_3}	G_{η_4}	G_{η_5}
A	3.267×10^{-1}	-1.711×10^0	4.210×10^{-2}	1.359×10^{-1}	8.249×10^{-2}
B	3.236×10^{-1}	-1.605×10^0	1.835×10^{-2}	-5.758×10^{-2}	-3.786×10^0
C	3.261×10^{-1}	-2.596×10^{-1}	3.770×10^{-2}	8.726×10^{-2}	-2.273×10^0
D	-5.428×10^{-3}	1.432×10^{-2}	-8.926×10^{-3}	-1.973×10^{-2}	-4.498×10^{-1}

194
 195

196 **Table 4.** Parameters needed to calculate the coefficients (G_{λ_i}) in Eq. (7) as a function of
 197 MW_{mixture} , HN/CN , and $w_{\text{iso-alkanes}}$ for the thermal conductivity ESCC model: An average
 198 MAPD of 2.2% is obtained for thermal conductivities using Eq. (7) for the mixture compositions
 199 used in the training set (134 data points).

G_{λ_i}	G_{λ_1}	G_{λ_2}	G_{λ_3}	G_{λ_4}
<i>A</i>	-4.798×10^{-1}	3.524×10^{-1}	-1.298×10^{-1}	7.313×10^{-1}
<i>B</i>	-2.261×10^{-2}	-2.183×10^{-2}	7.079×10^{-2}	-1.440×10^{-1}
<i>C</i>	5.153×10^{-1}	-2.431×10^{-1}	-2.523×10^{-2}	3.628×10^{-1}
<i>D</i>	1.894×10^{-1}	-7.804×10^{-2}	-1.064×10^{-2}	5.156×10^{-2}

200

201 Results and Discussion

202 Viscosities and thermal conductivities were predicted for the hydrocarbon mixtures and
 203 fuels in this study using the 2IP model, the ESCC model using Eqs. (6 and 7), the 3IP model, and
 204 two additional variations of the 2IP model. One variation of the 2IP model (referred to as 2IP-1Pfit)
 205 fits a single model parameter (coefficient *B* in the thermal conductivity model or *D* in the viscosity
 206 model) to minimize the average MAPD of the whole data set for each mixture. The second variation
 207 (referred to as 2IP-4Pfit) fits all four model parameters (*A*, *B*, *C*, and *D*) in Eq. (2) to minimize the
 208 average MAPD of the whole data set for each mixture and represents the best possible fit of Eq. (2)
 209 to the experimental data. The predictions using the 2IP, 2IP-1Pfit, and ESCC models are shown in
 210 the subsequent figures for selected mixtures. However, predictions for all available mixture data
 211 for the ESCC, 2IP, 3IP, 2IP-1Pfit, and 2IP-4Pfit models are provided in the supplementary
 212 information (SI).

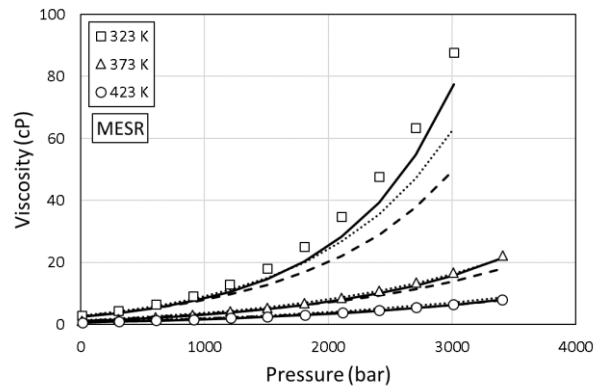
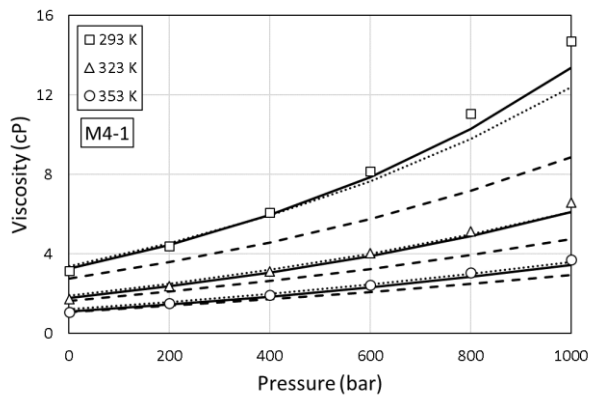
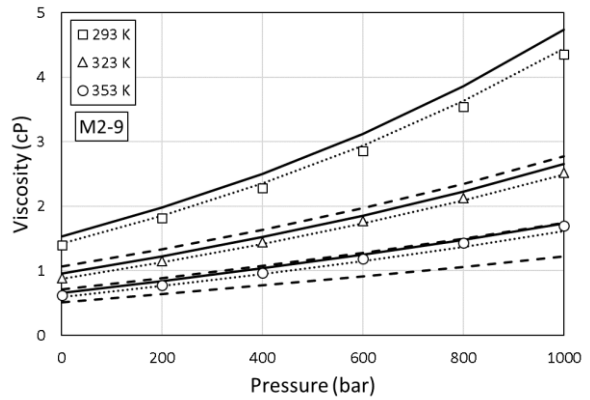
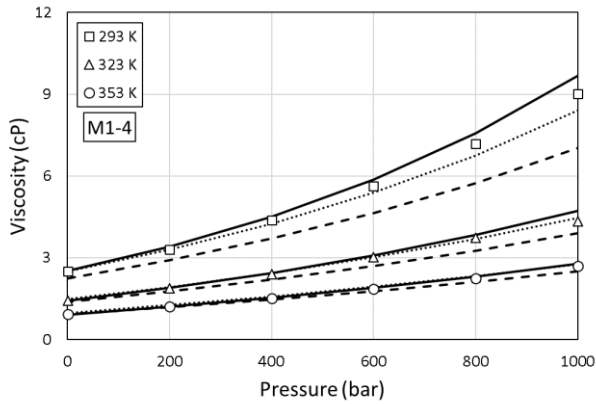
213 Figure 1 shows viscosity predictions using the 2IP, 2IP-1Pfit, and ESCC models in this
 214 study compared to experimental data for representative hydrocarbon mixtures and fuels, which
 215 contain a wide range of iso-alkane and two-ring saturate concentrations. M1-4 contains 55.1 wt%
 216 iso-alkanes, and the 2IP model predicts viscosity with 10.0% MAPD, whereas the 3IP, 2IP-1Pfit,

217 and ESCC models predict viscosity with MAPDs of 2.7, 2.6, and 2.3%, respectively. M2-9 contains
218 32.7 wt% iso-alkanes and 20.0 wt% two-ring saturates, and viscosity is predicted with 25.7%
219 MAPD using the 2IP model and 4.5% MAPD using the ESCC model. Viscosity predictions
220 improve over the ESCC model to 3.1 and 2.1% MAPD using the 3IP and 2IP-1Pfit models,
221 respectively but require experimental reference data to fit model parameters. M4-1 contains 80.6
222 wt% iso-alkanes, and the 2IP model predicts viscosity with 17.8% MAPD, whereas the 3IP, 2IP-
223 1Pfit, and ESCC models provide MAPDs of 8.1, 5.5, and 3.9%, respectively. The MESR diesel fuel
224 contains 27.3 wt% iso-alkanes and 4.8 wt% two-ring saturates, and the 2IP model predicts viscosity
225 with 13.4% MAPD. The 3IP and 2IP-1Pfit models slightly improve predictions to 11.6 and 11.1%
226 MAPD, respectively, while the ESCC model improves predictions further to 9.2% MAPD. Similar
227 observations are found for the other diesel fuels and mixtures. Table 5 compares the MAPDs for
228 viscosity predictions averaged over all mixtures and compositions using the ESCC model (4.8%),
229 2IP (15.0%), 3IP (5.5%), 2IP-1Pfit (4.1%), and 2IP-4Pfit (1.9%) models. The required input
230 parameters, as well as the MAPDs for viscosity predictions for all models, are reported in the SI
231 for all hydrocarbon mixture compositions and diesel fuels in this study.

232

233

234



235

236
237

238
239
240
241
242

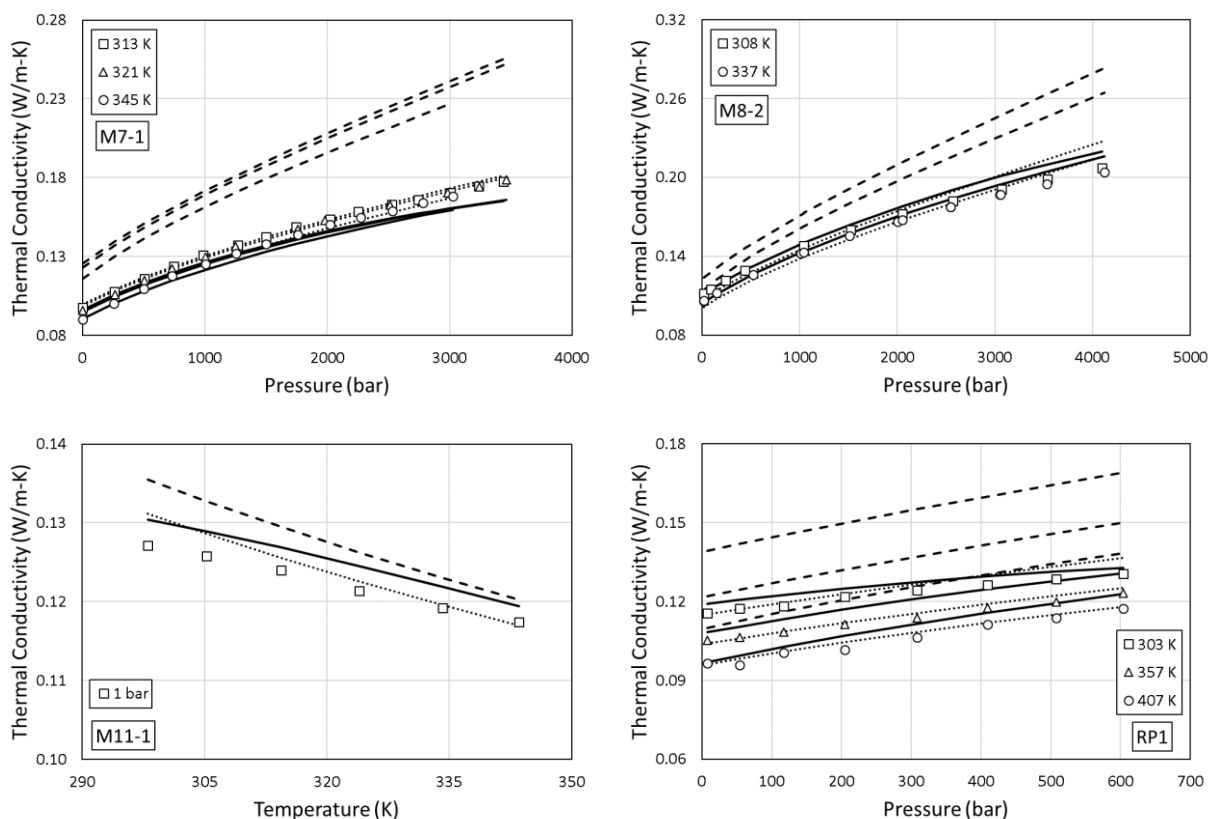
Figure 1. Pseudo-component viscosity predictions compared to experimental data [11, 37, 38, 40] (symbols) for selected hydrocarbon mixtures (M1-4, M2-9, and M4-1) and diesel fuel MESR using the 2IP (dashed lines), 2IP-1Pfit (dotted lines), and ESCC (solid lines) models. Results for the 3IP and 2IP-4IPfit models are listed in the SI but are not shown here to prevent cluttered graphs. Note that the y-axis and x-axis scales are different in each figure.

243 **Table 5.** The MAPD (%) for pseudo-component viscosity predictions using the 2IP, 3IP, 2IP-
 244 1Pfit, 2IP-4Pfit, and ESCC pseudo-component models.

Mixture or Fuel	2IP	2IP-4Pfit	3IP	2IP-1Pfit	ESCC
M1	13.9	1.8	3.6	5.6	3.6
M2	33.8	2.6	3.6	5.4	4.3
M3	5.3	2.3	2.9	3.0	6.1
M4	5.7	1.3	3.3	4.4	4.6
M5	7.1	1.9	2.3	3.4	7.0
M6	4.3	1.6	1.6	2.1	8.5
MESR	13.4	4.5	11.1	11.6	9.2
HNA	13.3	0.9	10.9	11.5	9.7
HPF	14.1	3.3	13.8	15.1	5.2
ULSD	12.2	3.8	8.3	9.5	7.0
HAR	12.9	2.6	12.2	12.3	5.4
Average	15.0	1.9	4.1	5.5	4.8

245
 246
 247 Figure 2 shows thermal conductivity predictions using the 2IP, 2IP-1Pfit, and ESCC
 248 models compared to the experimental data for representative hydrocarbon mixtures and fuels
 249 ranging from 0 to 100 wt% iso-alkanes. M7-1 contains 81.4 wt% iso-alkanes, and the 2IP model
 250 predicts thermal conductivity with 33.3% MAPD, whereas the 3IP, 2IP-1Pfit, and ESCC models
 251 provide MAPDs of 2.5, 1.1, and 3.9%, respectively. M8-2 contains 28.2 wt% iso-alkanes, and
 252 thermal conductivity is predicted with 18.4% MAPD using the 2IP model. Thermal conductivity
 253 predictions improve to 4.7 and 3.1% MAPD using the 3IP and 2IP-1Pfit models, but the ESCC
 254 model improves the MAPD further to 2.3%. M11-1 contains only normal alkanes, and the 2IP
 255 model predicts thermal conductivity with 4.4% MAPD. The 3IP and 2IP-1Pfit models provide
 256 slightly improved thermal conductivity predictions for M11-1 (1.6 and 1.4% MAPDs for 3IP and
 257 2IP-1Pfit models, respectively) compared to 2.3 % MAPD for the ESCC model. However, the 3IP
 258 and 2IP-1Pfit models require the input of experimental thermal conductivity reference data. The
 259 RP1 fuel contains 36.6 wt% iso-alkanes, and the 2IP model predicts thermal conductivity with
 260 20.4% MAPD. The 3IP and 2IP-1Pfit models improve predictions to 1.7 and 1.5% MAPD, while

261 the ESCC model predicts thermal conductivity with a 3.9% MAPD. Similar observations are found
 262 for other hydrocarbon mixtures and fuels. Table 6 lists the MAPDs for thermal conductivity
 263 predictions using the models for each hydrocarbon mixture (averaged over all compositions) and
 264 fuel. The MAPDs for thermal conductivity predictions averaged over all mixtures and
 265 compositions using the ESCC model (2.2%) are compared with the 2IP (9.3%), 3IP (2.4%), 2IP-
 266 1Pfit (1.6%), and 2IP-4Pfit (0.5%) models. The required input parameters, as well as the MAPDs
 267 for thermal conductivity predictions for all models, are reported in the SI for all hydrocarbon
 268 mixture compositions and fuels in this study.



269

270
271

Figure 2. Pseudo-component thermal conductivity predictions compared to experimental data [42-44, 46] (symbols) for selected hydrocarbon mixtures (M7-1, M8-2, and M11-1) and fuel RP1 using the 2IP (dashed lines), 2IP-1Pfit (dotted lines), and ESCC (solid lines) models. Results for the 3IP and 2IP-4IPfit models are listed in the SI but are not shown here to prevent cluttered graphs. Note that the y-axis and x-axis scales are different in each figure.

276 **Table 6.** The MAPD (%) for pseudo-component thermal conductivity predictions using the 2IP,
 277 3IP, 2IP-1Pfit, 2IP-4Pfit, and ESCC pseudo-component models.

Mixture or Fuel	2IP	2IP-4Pfit	3IP	2IP-1Pfit	ESCC
M7	24.5	0.9	0.9	1.7	2.2
M8	20.9	0.5	2.6	3.8	2.0
M9	1.6	0.3	1.2	1.9	0.8
M10	1.9	0.2	1.9	2.9	1.9
M11	3.2	0.4	1.3	2.2	1.2
M12	2.4	0.2	1.3	2.1	0.6
RP1	20.4	1.0	1.5	1.7	3.9
RP2	23.9	0.6	1.4	1.4	6.0
RP3	9.4	0.6	1.1	1.1	4.8
Jet A	14.6	0.6	2.1	3.4	4.4
JP-8 3773	21.3	0.8	1.0	2.4	5.8
Average	9.3	0.5	1.6	2.4	2.2

278

279

280 **Conclusion**

281 The two-input parameter (2IP) entropy-scaling based pseudo-component (ESBPC) model
 282 predicts viscosity and thermal conductivity accurately for many hydrocarbon mixtures and fuels
 283 (7 and 2% mean absolute percent deviation (MAPD) for viscosity and thermal conductivity,
 284 respectively, on average), but often less accurately when mixtures contain branched alkanes, with
 285 up to 24% mean absolute percent deviation (MAPD) calculated for fuels. A three-input parameter
 286 (3IP) version of the model improves predictions but requires fitting one model parameter using an
 287 experimental reference viscosity or thermal conductivity data point, which is not ideal if
 288 experimental reference data are not available. To enable more practical yet still accurate
 289 predictions, empirical correlations for the entropy-scaling based pseudo-component (ESBPC)
 290 coefficients were applied to the ESBPC technique to predict viscosity and thermal conductivity
 291 for hydrocarbon mixtures and fuels. Entropy is calculated in this Entropy Scaling Coefficient
 292 Corrected (ESCC) model using the pseudo-component technique. However, the viscosity and
 293 thermal conductivity coefficients are calculated using empirical correlations fit to a training set of

294 available high temperature and pressure literature data. The correlations for the thermal
295 conductivity pseudo-component coefficients require three mixture property inputs: the number
296 average molecular weight, hydrogen to carbon ratio, and the weight fraction of iso-alkanes. The
297 correlations for the viscosity pseudo-component correlations require four mixture property inputs:
298 the number averaged molecular weight, hydrogen to carbon ratio, the weight fraction of iso-
299 alkanes, and the weight fraction of two-ring saturates. Training sets of 690 and 134 points were
300 used to fit the correlation parameters for the viscosity and thermal conductivity models,
301 respectively, and a test set of 356 thermal conductivity and 1134 viscosity data points was used to
302 validate the correlations. The ESCC model does not require fitting model parameters to viscosity
303 or thermal conductivity reference data to predict viscosity and thermal conductivity up to high
304 pressures and temperatures, yet viscosities and thermal conductivities are predicted with 4.8 and
305 2.2% MAPD on average, respectively, which is comparable to the uncertainty of the experimental
306 data for the hydrocarbon mixtures (2%) and fuels (up to 4%) in this study.

307

308 **Acknowledgments**

309 This project has received funding from the European Union Horizon 2020 Research and
310 Innovation program, Grant Agreement No 675528. The authors wish to thank Ashutosh Gupta
311 (Afton), William B. Anderson (Afton), and Mark M^cHugh (Virginia Commonwealth University)
312 for their helpful and technical discussions.

313

314 **References**

- 315 [1] H.B. Rokni, Moore, J.D., Gupta, A., M^cHugh, M.A., Gavaises, M., Entropy scaling based viscosity
316 predictions for hydrocarbon mixtures and diesel fuels up to extreme conditions, *Fuel*, 241 (2019) 1203-
317 1213.
- 318 [2] H.B. Rokni, Moore, J.D., Gupta, A., M^cHugh, M.A., Mallepally, R.R., Gavaises, M., General method
319 for prediction of thermal conductivity for well-characterized hydrocarbon mixtures and fuels up to extreme
320 conditions using entropy scaling, *Fuel*, 245 (2019) 594-604.
- 321 [3] A. Theodorakakos, Strotos, G., Mitroglou, N., Atkin, C., Gavaises, M., Friction-induced heating in
322 nozzle hole micro-channels under extreme fuel pressurisation, *Fuel*, 123 (2014) 143-150.

323 [4] C. Rodriguez, Vidal, A., Koukouvinis, P., Gavaises, M., M^cHugh, M.A., Simulation of transcritical fluid
324 jets using the PC-SAFT EoS, *Journal of Computational Physics*, 374(1) (2018) 444-468.

325 [5] A. Parsa, Srinivasan, S., Saghir, M.Z., Impact of density gradients on the fluid flow inside a vibrating
326 cavity subjected to soot effect, *The Canadian Journal of Chemical Engineering*, 91(3) (2013) 550-559.

327 [6] C. Rodriguez, Vidal, A., Koukouvinis, P., Gavaises, M., Supercritical and transcritical real-fluid mixing
328 using the PC-SAFT EOS, in: *Ilass Europe, 28th Conference on Liquid Atomization and Spray Systems*,
329 Valencia, Spain, 2017, pp. 597-564.

330 [7] H. An, Yang, W., Li, J., Maghbouli, A., Chua, K.J., Chou, S.K., A numerical modeling on the emission
331 characteristics of a diesel engine fueled by diesel and biodiesel blend fuels, *Applied Energy*, 130 (2014)
332 458-465.

333 [8] B. Mohan, Yang, W., Yu, W., Effect of internal nozzle flow and thermo-physical properties on spray
334 characteristics of methyl esters, *Applied Energy*, 129 (2014) 123-134.

335 [9] A. Maghbouli, Yang, W., An, H., Li, J., Shafee, S., Effects of injection strategies and fuel injector
336 configuration on combustion and emission characteristics of a DI diesel engine fueled by bio-diesel,
337 *Renewable Energy*, 76 (2015) 687-698.

338 [10] P. Kontoulis, Kaiktsis, L., von Rotz, B., Boulouchos, K., CFD Modeling and Experimental Spray
339 Studies for Different Heavy Fuel Oil Qualities with Respect to Large Two-Stroke Marine Engines, *Journal*
340 *of Energy Engineering*, 145(5) (2019) 04019014.

341 [11] M. Aquino, Ciotta, F., Creton, B., Féjean, C., Pina, A., Dartiguelongue, C., Trusler, J.M., Vignais, R.,
342 Lugo, R., Ungerer, P., Composition analysis and viscosity prediction of complex fuel mixtures using a
343 molecular-based approach, *Energy & Fuels*, 26(4) (2012) 2220-2230.

344 [12] C. Schaschke, Fletcher, I., Glen, N., Density and viscosity measurement of diesel fuels at combined
345 high pressure and elevated temperature, *Processes*, 1(2) (2013) 30-48.

346 [13] F. Ramos-Pallares, Schoeggl, F.F., Taylor, S.D., Yarranton, H.W., Expanded fluid-based thermal
347 conductivity model for hydrocarbons and crude oils, *Fuel*, 224 (2018) 68-84.

348 [14] F. Ramos-Pallares, Schoeggl, F.F., Taylor, S.D., Yarranton, H.W., Prediction of thermal conductivity
349 for characterized oils and their fractions using an expanded fluid based model, *Fuel*, 234 (2018) 66-80.

350 [15] O.S. Alade, Ademodi, B., Sasaki, K., Sugai, Y., Kumasaka, J., Ogunlaja, A.S., Development of models
351 to predict the viscosity of a compressed Nigerian bitumen and rheological property of its emulsions, *Journal*
352 *of Petroleum Science and Engineering*, 145 (2016) 711-722.

353 [16] S. Eghbali, Dehghanpour, H., Dragani, J., Zhang, X., Phase Behaviour and Viscosity of Bitumen-CO
354 2/Light Hydrocarbon Mixtures at Elevated Temperatures: A Cold Lake Case Study, in: *SPE Canada Heavy*
355 *Oil Technical Conference*, Society of Petroleum Engineers, Calgary, Alberta, Canada, 2018.

356 [17] M. Gülüm, Bilgin, A., Measurements and empirical correlations in predicting biodiesel-diesel blends'
357 viscosity and density, *Fuel*, 199(567-77) (2017).

358 [18] S. Bair, The pressure and temperature dependence of volume and viscosity of four Diesel fuels, *Fuel*,
359 135 (2014) 112-119.

360 [19] S.O. Ilyin, Strelets, L.A., Basic Fundamentals of Petroleum Rheology and Their Application for the
361 Investigation of Crude Oils of Different Natures, *Energy & Fuels*, 32(1) (2018) 268-278.

362 [20] I.P. Kanaveli, Atzemi, M., Lois, E., Predicting the viscosity of diesel/biodiesel blends, *Fuel*, 199 (2017)
363 248-263.

364 [21] M. Lapuerta, Rodríguez-Fernández, J., Fernández-Rodríguez, D., Patiño-Camino, R., Modeling
365 viscosity of butanol and ethanol blends with diesel and biodiesel fuels, *Fuel*, 199 (2017) 332-338.

366 [22] N. Kolev, *Multiphase Flow Dynamics 3: Turbulence, Gas Absorption and Release, Diesel Fuel*
367 *Properties*. **2002**, in, Springer Verlag Berlin Heidelberg.

368 [23] H. Baled, Gamwo, I.K., Enick, R.M., M^cHugh, M.A., Viscosity models for pure hydrocarbons at
369 extreme conditions: A review and comparative study, *Fuel*, 218 (**2018**) 89-111.

370 [24] H.B. Rokni, Gupta, A., Moore, J.D., M^cHugh, M.A., Bamgbade, B.A., Gavaises, M., Purely predictive
371 method for density, compressibility, and expansivity for hydrocarbon mixtures and diesel and jet fuels up
372 to high temperatures and pressures, *Fuel*, 236 (**2019**) 1377-1390.

373 [25] H.W. Yarranton, Satyro, M.A., Expanded fluid-based viscosity correlation for hydrocarbons, *Industrial*
374 *& Engineering Chemistry Research*, 48(7) (**2009**) 3640-3648.

375 [26] H. Motahhari, Satyro, M.A., Taylor, S.D., Yarranton, H.W., Extension of the expanded fluid viscosity
376 model to characterized oils, *Energy & Fuels*, 27(4) (**2013**) 1881-1898.

377 [27] H. Motahhari, Schoeggl, F., Satyro, M., Yarranton, H., Viscosity prediction for solvent-diluted live
378 bitumen and heavy oil at temperatures up to 175-deg-C, *Journal of Canadian Petroleum Technology*, 52(5)
379 (**2013**) 376-390.

380 [28] M. Ma, Chen, S., Abedi, J., Modeling the density, solubility and viscosity of bitumen/solvent systems
381 using PC-SAFT, *Journal of Petroleum Science and Engineering*, 139 (**2016**) 1-12.

382 [29] S.E. Quiñones-Cisneros, Zéberg-Mikkelsen, C.K., Stenby, E.H., The friction theory (f-theory) for
383 viscosity modeling, *Fluid Phase Equilibria*, 169(2) (**2000**) 249-276.

384 [30] S.E. Quiñones-Cisneros, Zéberg-Mikkelsen, C.K., Baylaucq, A., Boned, C., Viscosity modeling and
385 prediction of reservoir fluids: From natural gas to heavy oils, *International Journal of Thermophysics*, 25(5)
386 (**2004**) 1353-1366.

387 [31] S.E. Quiñones-Cisneros, Dalberg, A., Stenby, E.H., PVT characterization and viscosity modeling and
388 prediction of crude oils, *Petroleum Science and Technology*, 22((9-10)) (**2004**) 1309-1325.

389 [32] K.A. Schmidt, Quiñones-Cisneros, S.E., Kvamme, B., Density and viscosity behavior of a North Sea
390 crude oil, natural gas liquid, and their mixtures, *Energy & Fuels*, 19(4) (**2005**) 1303-1313.

391 [33] M.I. Abutaqiya, Zhang, J., Vargas, F.M., Viscosity modeling of reservoir fluids using the Friction
392 Theory with PC-SAFT crude oil characterization, *Fuel*, 235 (**2019**) 113-129.

393 [34] Y. Khoshnamvand, Assareh, M., Viscosity Prediction for Petroleum Fluids Using Free Volume Theory
394 and PC-SAFT, *International Journal of Thermophysics*, 39(4) (**2018**) 54.

395 [35] Y. Sun, Shen, G., Held, C., Lu, X., Ji, X., Modeling Viscosity of Ionic Liquids with Electrolyte
396 Perturbed-Chain Statistical Associating Fluid Theory and Free Volume Theory, *Industrial & Engineering*
397 *Chemistry Research*, 57(26) (**2018**) 8784-8801.

398 [36] J. Gross, Sadowski, G., Perturbed-chain SAFT: An equation of state based on a perturbation theory for
399 chain molecules, *Industrial & Engineering Chemistry Research*, 40(4) (**2001**) 1244-1260.

400 [37] P. Dauge, Canet, X., Baylaucq, A., Boned, C., Measurements of the density and viscosity of the
401 tridecane+ 2, 2, 4, 4, 6, 8, 8-heptamethylnonane mixtures in the temperature range 293.15-353.15 K at
402 pressures up to 100 MPa, *High Temperatures High Pressures*, 33(2) (**2001**) 213-230.

403 [38] C.K. Zéberg-Mikkelsen, Barrouhou, M., Baylaucq, A., Boned, C., High-pressure viscosity and density
404 measurements of the ternary system methylcyclohexane+ cis-decalin+ 2, 2, 4, 4, 6, 8, 8-heptamethylnonane,
405 *Journal of Chemical & Engineering Data*, 48(6) (**2003**) 1387-1392.

406 [39] D. Ducoulombier, Zhou, H., Boned, C., Peyrelasse, J., Saint-Guirons, H., Xans, P., Pressure (1-1000
407 bars) and temperature (20-100. degree. C) dependence of the viscosity of liquid hydrocarbons, *The Journal*
408 *of Physical Chemistry*, 90(8) (**1986**) 1692-1700.

409 [40] C.K. Zeberg-Mikkelsen, Canet, X., Baylaucq, A., Quiñones-Cisneros, S.E., Boned, C., Stenby, E.H.,
410 High-pressure viscosity and density behavior of ternary mixtures: 1-methylnaphthalene+ n-tridecane+ 2, 2,
411 4, 4, 6, 8, 8-heptamethylnonane, *International Journal of Thermophysics*, 22(6) (2001) 1691-1726.

412 [41] C. Boned, Zeberg-Mikkelsen, C.K., Baylaucq, A., Daugé, P., High-pressure dynamic viscosity and
413 density of two synthetic hydrocarbon mixtures representative of some heavy petroleum distillation cuts,
414 *Fluid Phase Equilibria*, 212(1) (2003) 143-164.

415 [42] W.A. Wakeham, Yu, H.R., Zalaf, M., The thermal conductivity of the mixtures of liquid hydrocarbons
416 at pressures up to 400 MPa, *International Journal of Thermophysics*, 11(6) (1990) 987-1000.

417 [43] J.M.N.A. Fareleira, Li, S.F.Y., Wakeham, W.A., The thermal conductivity of liquid mixtures at
418 elevated pressures, *International Journal of Thermophysics*, 10(5) (1989) 1041-1051.

419 [44] Y. Wada, Nagasaka, Y., Nagashima, A., Measurements and correlation of the thermal conductivity of
420 liquid n-paraffin hydrocarbons and their binary and ternary mixtures, *International Journal of*
421 *Thermophysics*, 6(3) (1985) 251-265.

422 [45] A.J. Rowane, Mahesh, B.V., Rokni, H.B., Moore, J.D., Gavaises, M., Wensing, M., Gupta, A.,
423 M'Hugh, M.A., Effect of Composition, Temperature, and Pressure on the Viscosities and Densities of Three
424 Diesel Fuels, *Journal of Chemical & Engineering Data*, 64(12) (2019) 5529-5547.

425 [46] L.A. Akhmedova-Azizova, Abdulagatov, I.M., Bruno, T.J., Effect of RP-1 compositional variability
426 on thermal conductivity at high temperatures and high pressures, *Energy & Fuels*, 23(9) (2009) 4522-4528.

427 [47] T.J. Bruno, The Properties of RP-1 and RP-2 MIPR F1SBAA8022G001, 2008.

428 [48] G.Q. Xu, Jia, Z.X., Wen, J., Deng, H.W., Fu, Y.C., Thermal-conductivity measurements of aviation
429 kerosene RP-3 from (285 to 513) K at sub-and supercritical pressures, *International Journal of*
430 *Thermophysics*, 36(4) (2015) 620-632.

431 [49] Z. Jia, Xu, G., Deng, H., Jie, W., Fu, Y., Experimental measurements of thermal conductivity of
432 hydrocarbon fuels by a steady and kinetic method, *Journal of Thermal Analysis and Calorimetry*, 123(1)
433 (2016) 891-898.

434 [50] T.J. Bruno, Thermodynamic, Transport and Chemical Properties of Reference JP-8, NIST, 2006.

435 [51] R. Xu, Wang, K., Banerjee, S., Shao, J., Parise, T., Zhu, Y., Wang, S., Movaghar, A., Lee, D.J., Zhao,
436 R., Han, X., Gao, Y., Lu, T., Brezinsky, K., Egolfopoulos, F.N., Davidson, D.F., Hanson, R.K., Bowman,
437 C.T., Wang, H., A physics-based approach to modeling real-fuel combustion chemistry – II. Reaction
438 kinetic models of jet and rocket fuels, *Combustion and Flame*, 193 (2018) 520-537.

439 [52] J.T. Edwards, Reference jet fuels for combustion testing, in: 55th AIAA Aerospace Sciences Meeting,
440 2017, pp. 0146.

441 [53] Y. Rosenfeld, Relation between the transport coefficients and the internal entropy of simple systems,
442 *Physical Review A*, 15(6) (1977).

443 [54] O. Lötgering-Lin, J. Gross, Group contribution method for viscosities based on entropy scaling using
444 the perturbed-chain polar statistical associating fluid theory, *Industrial & Engineering Chemistry Research*,
445 54(32) (2015) 7942-7952.

446 [55] L. Novak, Self-diffusion coefficient and viscosity in fluids, *Journal of Chemical Reactor Engineering*,
447 9(1) (2011).

448 [56] L. Novak, Fluid viscosity-residual entropy correlation, *International Journal of Chemical Reactor*
449 *Engineering*, 9(1) (2011).

450 [57] M. Hopp, Gross, J., Thermal Conductivity of Real Substances from Excess Entropy Scaling Using
451 PCP-SAFT, *Industrial & Engineering Chemistry Research*, 56(15) (2017) 4527-4538.

452