A Residual Entropy Scaling Approach for Viscosity of Refrigerants, Other Fluids and their Mixtures

Xiaoxian YANG^{*(a,b)}, Xiong XIAO^(b), Monika THOL^(c), Markus RICHTER^(a,b) and Ian H. BELL^(d)

 ^(a) Applied Thermodynamics, Chemnitz University of Technology Chemnitz, 09107, Germany
 ^(b) Fluid Science & Resources Division, School of Engineering, University of Western Australia Crawley, WA 6009, Australia
 ^(c) Thermodynamics, Ruhr University Bochum Bochum, 44780, Germany
 ^(d) Applied Chemicals and Materials Division, National Institute of Standards and Technology Boulder, Colorado 80305, United States
 *Corresponding author: xiaoxian.yang@mb.tu-chemnitz.de

ABSTRACT

In our previous work (*J. Chem. Eng. Data* 2021, 66, 3, 1385–1398), a residual entropy scaling (RES) approach was developed to link viscosity to residual entropy using a simple polynomial equation for refrigerants. The present study extends the RES approach to other fluids and their mixtures with refrigerants. More than 68.2% (one standard deviation) of the well-selected experimental data agree with the RES model within 3.2% and 8.0% for 124 pure fluids and their mixtures, respectively. With advantageously many fewer parameters, the RES approach yields similar statistical agreement with the experimental data as the best-selected models implemented in the NIST REFPROP database, the current state-of-the-art for thermophysical property calculations. The present work is the basis for developing a RES approach for mixtures of refrigerants with lubricants relevant to compressor design. This new viscosity model will be implemented in an open source software package.

Keywords: Entropy Scaling, Mixture, Multi-Parameter Equation of State, Refrigerant, Thermophysical Property Modelling, Viscosity

1. INTRODUCTION

Refrigerants, as working fluids, and lubricants, required to run many types of compressors, both play important roles in many industrial applications, such as refrigeration and power generation. Reliable knowledge of thermophysical properties of refrigerant-lubricant mixtures is essential to evaluate the performance of refrigeration and energy systems. However, there are virtually no reliable models for accurate property calculations. To tackle this issue, the present work extends the residual entropy scaling (RES) approach for viscosity developed in our previous work (Yang et al. 2021c) from refrigerants to all pure fluids whose reference equation of state (EoS) are available in the NIST REFPROP database 10.0 (Lemmon et al. 2018) and whose experimental viscosity data are available to us. This endeavor is part of the research program within the KETEC project (Forschungsplattform Kälte- und Energietechnik), a research platform for the next-generation refrigeration and energy technology funded by Federal Ministry of Education and Research of Germany since 2021 (see details at http://ketec.online). One important goal within subproject 3 of the KETEC project is to make the RES approach applicable for refrigerant + lubricant mixtures. The core part of the current research has been published open access as a peer-reviewed article (Yang et al. 2022).

2. THEORETICAL BACKGROUND

The RES approach expresses transport properties in terms of residual entropy, which can be obtained directly from an EoS. Various approaches based on RES have been proposed and verified for viscosity of the Lennard-Jones fluid (Bell et al. 2019) and hundreds of real fluids, e.g., hydrocarbons (Al Ghafri et al. 2021; Bell 2020a,b; Binti Mohd Taib & Trusler 2020), refrigerants (Bell & Laesecke 2016; Liu et al. 2020; Wang et al. 2020; Yang et al. 2021c,b), or other commonly used fluids (Bell 2019; Lötgering-Lin et al. 2018), as well as thermal conductivity of some real fluids (Fouad 2020; Hopp et al. 2019; Hopp & Gross 2017; Kim et al. 2021; Liu et al. 2021; Wang et al. 2021a). Here the approach developed in our previous work (Yang et al. 2021c) is extended and the mixing rule is slightly modified.

The fluid viscosity η is calculated as the sum of the dilute gas viscosity $\eta_{\rho \to 0}(T)$ and the residual part $\eta_{res}(s^r)$:

$$\eta = \eta_{\rho \to 0}(T) + \eta_{\text{res}}(s^{r})$$
Eq. (1)

The $\eta_{\rho \to 0}(T)$ at temperature *T* of a pure fluid is calculated with the Chapman-Enskog (Hirschfelder et al. 1964) solution of the Boltzmann transport equation, assuming the interactions between molecules can be roughly captured by those of Lennard-Jones (L-J) particles with 12-6 potential:

$$\eta_{\rho \to 0}(T) = \frac{5}{16} \sqrt{\frac{mk_{\rm B}T}{\pi}} \frac{1}{\sigma^2 \Omega^{(2,2)*}}$$
 Eq. (2)

where *m*, in units of kg is the mass of one molecule; $k_{\rm B} = 1.380649 \cdot 10^{-23} \text{ J} \cdot \text{K}^{-1}$ is the Boltzmann constant; σ is the collision diameter of the L-J particle; and $\Omega^{(2,2)^*}$ is the reduced collision integral obtained by integrating the possible approach trajectories of the particles. (Neufeld et al. 1972) gives an empirical correlation of $\Omega^{(2,2)^*}$ as a function of temperature as:

$$\Omega^{(2,2)*} = 1.16145 \cdot (T^*)^{-0.14874} + 0.52487 \cdot \exp(-0.77320 \cdot T^*) + 2.16178 \qquad \text{Eq. (3)} \\ \cdot \exp(-2.43787 \cdot T^*)$$

where $T^* = k_B T/\varepsilon$ is the dimensionless temperature, and ε/k_B is the reduced L-J pair-potential energy. The non-polynomial terms are neglected in this work as REFPROP 10.0 does. The L-J parameters (σ and ε) in this work were obtained from REFPROP 10.0.

As introduced by (Bell 2019, 2020a), the residual part of viscosity $\eta_{res}(s^r)$ can be calculated with:

$$\eta_{\rm res}(s^{\rm r}) = \frac{\eta_{\rm res}^{+} \rho_{\rm N}^{2/3} \sqrt{mk_{\rm B}T}}{(s^{+})^{2/3}}$$
 Eq. (4)

$$s^+ = -s^r/R$$
 Eq. (5)

Here, ρ_N , in units of m⁻³, is the number density; s^r in units of J·mol⁻¹·K⁻¹ is the molar residual entropy, defined as the difference between the real fluid entropy and the ideal gas entropy at the same temperature and density; and $R = 8.31446261815324 \text{ J·mol}^{-1} \cdot \text{K}^{-1}$ is the molar gas constant (Tiesinga et al. 2021). In this work, the number density ρ_N and molar residual entropy s^r were calculated with the reference EoS implemented in REFPROP 10.0 (Lemmon et al. 2018) using the python CoolProp package 6.4.1 (Bell et al. 2014) as an interface. The plus-scaled dimensionless residual viscosity η_{res}^+ is related to the plus-scaled dimensionless residual entropy s^+ using the following polynomial equations

$$\ln(\eta_{\rm res}^+ + 1) = n_1 \cdot (s^+) + n_2 \cdot (s^+)^{1.5} + n_3 \cdot (s^+)^2 + n_4 \cdot (s^+)^{2.5}$$
 Eq. (6)

or

$$\ln(\eta_{\rm res}^+ + 1) = n_{\rm g1} \cdot (s^+/\xi) + n_{\rm g2} \cdot (s^+/\xi)^{1.5} + n_{\rm g3} \cdot (s^+/\xi)^2 + n_{\rm g4} \cdot (s^+/\xi)^{2.5}$$
 Eq. (7)

Eq. (6) is for a pure fluid with fluid-specific fitted parameter n_k (k = 1, 2, 3, 4), and Eq. (7) is for a group of pure fluids with global fitted parameters n_{gk} (k = 1, 2, 3, 4) and a fluid-specific scaling factor ξ for each pure fluid.

To extend the RES model to mixtures, a predictive mixing rule is adopted. The dilute gas viscosity $\eta_{\rho \to 0, \text{mix}}$ is calculated with the approximation of (Wilke 1950):

$$\eta_{\rho \to 0, \text{mix}} = \sum_{i=1}^{N} \frac{x_i \cdot \eta_{\rho \to 0, i}}{\sum_{j=1}^{N} x_j \cdot \varphi_{ij}}$$
Eq. (8)

with

$$\varphi_{ij} = \frac{\left(1 + (\eta_{\rho \to 0,i}/\eta_{\rho \to 0,j})^{1/2} \cdot (m_j/m_i)^{1/4}\right)^2}{(8 \cdot (1 + m_i/m_j))^{1/2}},$$
 Eq. (9)

where x_i is the mole fraction of component *i* and m_i is the mass of one molecule of component *i*. The mole fraction weighted average m_{mix} of the components is used to replace the effective mass of one particle *m* in Eq. (4):

$$m_{\rm mix} = \sum_{i} x_i \cdot m_i$$
 Eq. (10)

Attempts to use a mass fraction weighted average result in a negligible statistical difference. Then, in contrast to our previous work (Yang et al. 2021c), the mole fraction weighted average coefficient $n_{k,mix}$ is utilised to substitute the parameters n_k in Eq. (6), i.e.,

$$n_{k,\text{mix}} = \sum_{i} x_i \cdot n_{k,i}$$
 Eq. (11)

where $n_{k,i}$ (k = 1,2,3,4) are fitted n_k parameters of component *i*. It is important to note that, only if a pure fluid does not have fluid-specific fitted parameters, the n_k (k = 1, 2, 3, 4) are replaced by n_{g1}/ξ , $n_{g2}/\xi^{1.5}$, n_{g3}/ξ^2 , and $n_{g4}/\xi^{2.5}$, respectively.

3. RESULTS

In total, 51841 experimental (T, p, η) values of 124 pure fluids and 33036 experimental (x, T, p, η) values of 351 mixtures were collected. These experimental data were obtained from approximately 1846 literature sources (Yang et al. 2022) mainly from NIST's ThermoData Engine (TDE) (Frenkel et al. 2005) database. The same method as in our previous work (Yang et al. 2021c,a) to correct a small portion of data from the TDE database (less than 0.1 %, mainly due to mistakes in data transfer from original sources to the database), and the same filters to sort out inappropriate data were carried out. In total, 6.2 % of pure-fluid and 8.9 % of mixture data were filtered out.

3.1. Correlation for pure fluids

At first, the fluid-specific n_k parameters in Eq. (6) were obtained for those pure fluids with a sufficient quantity and good quality of experimental data in both liquid and gas phases. The results are listed in Table 1 in the published peer-reviewed article (Yang et al. 2022). The method to fit the n_k parameters as well as the global $n_{g,k}$ parameters to be discussed below is described in our previous work (Yang et al. 2021c). Then, the classification of the 124 pure fluids was carried out to achieve the goal of the RES model with the global $n_{g,k}$ parameters and the fluid-specific scaling factor ξ having the best statistical agreement with experimental data for each pure fluid while keeping the number of groups as small as possible. Ultimately, the 124 pure fluids were classified into eight groups, as illustrated in Figure 1. From groups 1 to 8, the fluids are mainly but not exactly: (1-LG) light gases with quantum effects at low temperatures, mainly hydrogen and its spin isomers and helium; (2-G) gaseous fluids, e.g., the noble gases; (3-LHC) a majority of light hydrocarbons and halogenated hydrocarbons (refrigerants); (4-B) fluids with benzene rings and similar fluids; (5-MHC) medium hydrocarbons and similar fluids; (6-HHC) heavy hydrocarbons and dense fluids; (7-LA) fluids with light intermolecular association among molecules like methanol; (8-SA) fluids with strong intermolecular association among molecules, such as water. The global $n_{g,k}$ parameters for each group are listed in Table 2. Experimental data of each group collapse into the individual global η_{res}^+ vs. s^+ curves as shown in Figure 2.



Figure 1. Scaling factor ξ . The denominator s^+_{crit} is the plus-scaled dimensionless residual entropy at the critical point calculated with REFPROP 10.0 (Lemmon et al. 2018) for each pure fluid. The number at the top right of each box indicates the group number. The vertical dashed dotted line denotes $\xi / s^+_{crit} = 0.7$. The fluid names were taken as they are used in REFPROP 10.0. This figure is taken, unmodified from the journal paper (Yang et al. 2022).

Table 1. Global fitted parameters of each group					
Group	n _{g1}	<i>n</i> _{g2}	n _{g3}	n _{g4}	
1	-0.449854	3.219854	-5.298638	2.975827	
2	0.101609	-0.156239	0.440241	-0.113646	
3	-0.448046	1.012681	-0.381869	0.054674	
4	-0.657607	1.154900	-0.437437	0.059896	
5	-0.368714	0.764423	-0.261237	0.031913	
6	0.645294	-0.186122	0.041308	-0.002865	
7	-0.726184	1.369572	-0.652048	0.116681	
8	-0.663915	1.401829	-0.780113	0.155976	



Figure 2. Values of $(\eta_{res}^{+} + 1)$ as a function of s^{+}/ξ for each group of pure fluids, where is η_{res}^{+} the plus-scaled dimensionless residual viscosity, s^{+} is the plus-scaled residual entropy, and ξ is the scaling factor. The curves are calculated with the global n_{gk} parameters. All groups are shown at the bottom; at the top, each group is individually illustrated but stacked by powers of 20 and with group number labeled. This figure is taken, unmodified from the journal paper (Yang et al. 2022).

According to Figure 1, there is a relation between ξ and s^+_{crit} , the plus-scaled dimensionless residual entropy at the critical point. For example, ξ / s^+_{crit} is roughly a value of 0.7 in group 3, and for a group with heavier components, the value of ξ / s^+_{crit} decreases. This factor is in good agreement with the scaling shown by (Bell 2020b; Bell et al. 2021). Adopting the group classification and the average value of ξ / s^+_{crit} for that group, the RES model could serve as a fully predictive model for other chemically similar pure fluids.

A summary of the relative deviations of the experimental viscosity η_{exp} from values η_{RES} calculated with the RES model is shown in Figure 3 for refrigerants; similar figures for more fluids are available in the published article (Yang et al. 2022). It is important to note again, fluid-specific n_k parameters are preferred in all calculations in this work, and only if they are not available, global parameters n_{gk} are used. As a result, more

than 68.2% of the experimental data (corresponding to one standard deviation) agree with the RES model within 3.2%.

Here we defined average relative deviation (ARD) and average of the absolute value of relative deviation (AARD) of the experimental values η_{exp} from the model calculations η_{RES} as:

$$ARD = \frac{\sum_{i}^{N} [(\eta_{exp,i} - \eta_{RES,i})/\eta_{RES,i}]}{N}$$
(12)

$$AARD = \frac{\sum_{i}^{N} |(\eta_{\exp,i} - \eta_{\text{RES},i})/\eta_{\text{RES},i}|}{N}$$
(13)

where *N* is the total number of the experimental data points for a given fluid. The values of ARD and AARD denote the systematic offset and scatter, respectively, of the experimental data from the model. The ARD for each pure fluid are shown in Figure 3. Ideally, ARD should be approximately zero for pure fluids as the RES model for pure fluids is anchored to the experimental data. However, considering the existence of low-quality data and the possible uncertainties due to the modeling of the dilute gas, the absolute value of ARD for only 91 and 113 of the 124 pure fluids are less than 1.0% and 2.0%, respectively. For those with larger ARD, there are either only gas phase data available (e.g., R116 and R161), conflicting datasets (see the gourd-shaped pattern for e.g., ammonia and n-butane in Figure 3), obviously inaccurate calculation of dilute gas viscosity or very few experimental data available. For pure fluids for which more than 1000 experimental data points are available, the absolute ARD values are generally less than 1.0% and all are less than 2.0%.



Figure 3. Relative deviations of the experimental viscosity η_{exp} from values η_{RES} calculated with the RES model. The short line indicates the average relative deviation; the shape shows the distribution of the relative deviation; and the colors are for a clear illustration only. Fluid-specific n_k parameters are preferred, and only if they are not available in

Table 1, global parameters n_{gk}/ξ are used. The fluid names were taken as they are used in REFPROP 10.0. This figure is taken, unmodified from the journal paper (Yang et al. 2021c).

We compared the performance of the RES model with the recommended model of each pure fluid implemented in the REFPROP 10.0, which we will refer to as 'REFPROP-models'. The REFPROP-models are 41 reference correlations for 43 pure fluids, the extended corresponding states (ECS) model for 77 pure fluids, and the friction theory model for 3 pure fluids. There is no viscosity model for NF₃ in REFPROP 10.0. The REFPROP-models fail in the calculation of very few experimental data (less than 0.38%, exceeding the model limit) at the given temperature and pressure. The RES model yields smaller or equal AARD (i.e., smaller

scatter) for 55 pure fluids out of 124 fluids compared to the REFPROP-models; this value becomes 61 out of 124 if the dilute gas viscosity in the RES model is calculated in the same way as the REFPROP-models do (achieved by setting pressure zero using the recommended models in REFPROP). In summary, the RES approach could yield similar statistical agreement with the experimental data as the state-of-the-art REFPROP-models, which have much more complicated formulation and more parameters.

Additional comparisons were made to the RES approach developed by (Lötgering-Lin et al. 2018), where the residual entropy is calculated with the PCP-SAFT EoS (Gross & Sadowski 2001). The calculations with the PCP-SAFT EoS were carried out with the TREND 5.0 package (Span et al. 2020) and the model parameters were obtained from the supporting information of (Lötgering-Lin et al. 2018) There are 35 pure fluids for which all three models (the REFPROP-models are considered as one model here) can be applied. For a fair comparison, approximately 6 % of the evaluated experimental data were further filtered out; these are mainly near the phase boundaries as the PCP-SAFT EoS predicts different phase boundaries than the multi-parameter reference EoS. Relative deviations of the experimental data from the three models are statistically shown in Figure 4. The ARD for the 35 fluids are shown in Figure 4. Our RES model yields the smallest AARD (i.e., smallest scatter) for 11 pure fluids while that for model of (Lötgering-Lin et al. 2018) is best for 3 fluids.



Figure 4. Relative deviations of the experimental viscosity η_{exp} of pure fluids from values η_{RES} calculated with the RES model, REFPROP models and model of (Lötgering-Lin et al. 2018). The short line indicates the average relative deviation; the shape shows the distribution of the relative deviation; and the colors are for a clear illustration only. Relative deviations of the experimental data of ethanol and methanol from Lötgering-Lin et al. model exceed the figure limits (with ARD more than 30 %). The fluid names were taken as they are used in REFPROP 10.0. This figure is taken, unmodified from the journal paper (Yang et al. 2022).

3.2. Prediction for mixtures

For mixtures, a predictive mixing rule was used, see section 2. More than 68.2% of the evaluated experimental data agree with the RES model within 8%. A summary is provided in Figure 5 showing the ARD (systematic offset) and AARD (scatter) from the experimental data to the RES model for binaries among two groups. For binaries from the same group, the absolute value of ARD is generally less than 2%; in particular, the ARD is only -0.4% for binaries within group 3 (4856 experimental data). For binaries from different groups, some are very good, e.g., 1982 data from groups 1 and 2 having an ARD of -1.2%, while some are relatively poor, e.g., 3368 data from groups 7 and 8 with an ARD of 17%. There seem to be problems in the EoS for the residual entropy calculations of binaries from groups 7 and 8 (e.g., Ethanol +Water), as will be discussed in the next paragraph, the REFPROP-models which also rely on the EoS fails in most of the calculations for

binaries from groups 7 and 8. Regarding asymmetric mixtures of industrial interest, such as refrigerants with lubricants (respectively in group 3 and possibly group 6), and hydrogen with heavy hydrocarbon (groups 1 and 5 or 6) (Thol & Richter 2021), very few experimental data are available. Therefore, Figure 5 reveals an AARD clearly beyond 10% for groups 3 and 6, and for groups 1 and 5 or 6, the AARD cannot be calculated (no data available).

We first compared the performance of the RES model and REFPROP-models in mixture prediction. Please note, there are up to four additional binary interaction parameters for each binary in the ECS model (the most commonly adopted model in REFPROP-models), and these parameters are fitted to the available experimental data or otherwise are set to zero. The REFPROP-models fail to calculate 21 % of the evaluated experimental data at the given temperature and pressure, mainly belonging to binaries including group 7 and group 8. After removing these data, statistical results compared to the experimental data are shown in the bottom line of Figure 5. The RES model yields lower AARD (scatter) for 161 mixtures out of all 351 mixtures; this value is 185 out of 351 if the dilute gas viscosity in the RES model is calculated in the same way as the REFPROP-models do. According to Figure 5, the RES model yields lower AARD (scatter) for 12 group pairs out of 18 group pairs where experimental data are available.



Figure 5. Statistical summary of the relative deviation of the experimental data from model calculations for binary mixtures. Top row: all evaluated experimental data; Bottom row: evaluated experimental data were further filtered for the calculations using the REFPROP-models. ARD: average relative deviation, and AARD: average of the absolute value of relative deviation, of the experimental values from the model calculations. Please note, for those without available data, ARD and ARRD are given a value of 0.0. The shading corresponds to the magnitude of the entries in the cells. This figure is taken, unmodified from the journal paper (Yang et al. 2022).

We then added the model of (Lötgering-Lin et al. 2018) into the comparison. Considering that there are only 35 pure fluids for which all three models (the REFPROP-models are considered as one model here) can be applied for, the binary mixtures were narrowed down to only 158. For a fairer comparison, approximately 2.8 % of the evaluated experimental data at or near phase boundaries was further filtered out as the PCP-SAFT and the multi-parameter reference EoS predict different phase boundaries. The statistical summary of the comparison from experimental data to the three models are illustrated for random-selected mixtures in Figure 6; similar figures for more fluids are available in the published article (Yang et al. 2022). Our RES model, the REFPROP-models, and the model of (Lötgering-Lin et al. 2018) have the best agreement with

experimental data for 56, 69 and 33 mixtures, respectively. It is interesting to note that, for some binary mixtures, such as *n*-pentane + toluene, nonane + *n*-pentane, *n*-hexane + *p*-xylene, and decane + *p*-xylene (see figures in the SI-DPR) experimental data in the liquid phase have similar deviations with all three models, i.e., the models agree with each other while the experimental data deviate.



Figure 6. Relative deviations of the experimental viscosity η_{exp} of selected mixtures from values η_{RES} calculated with the RES model, REFPROP-models and model of (Lötgering-Lin et al. 2018). The short line indicates the average relative deviation; the shape shows the distribution of the relative deviation; and the colors are for a clear illustration only. This figure is taken, unmodified from the journal paper (Yang et al. 2022).

4. CONCLUSION

In this work, we present a simple but accurate residual entropy scaling (RES) approach for all 124 pure fluids whose reference EoS (implemented in the NIST REFPROP database 10.0) and experimental viscosity data are available. More than 68.2% (corresponding to one standard deviation) of the evaluated experimental data agree with the RES model within 3.2% for pure fluids. The pure fluids are classified into 8 groups, and fluids with similar physical properties are roughly in the same group. Experimental data of each group collapse into a global residual viscosity vs. scaled residual entropy curve. There is a relation between the fluid-specific scaling factor and the plus-scaled dimensionless residual entropy at the critical point. According to this and adopting the classification, the RES model could serve as a fully predictive model for other pure fluids. Please note: fluid-specific fitted parameters should be used, and only if they are not available, global fitted parameters are used.

Compared to the recommended models implemented in the REFPROP 10.0, which is considered the stateof-the-art for thermophysical property calculation, the RES model yields smaller or equal average of the absolute value of the relative deviation (AARD) from the experimental data for 55 pure fluids out of 124 fluids. If the dilute gas viscosity in the RES model is calculated in the same way as the REFPROP-models do, this value becomes 61 out of 124. Future developments in the reference EoS, which improve the accuracy in the residual entropy calculation, should significantly improve the accuracy of the RES model, mainly in the liquid phase. With a predictive mixing rule, more than 68.2% of the evaluated experimental mixture data agree with the RES model within 8%. With more sophisticated mixing rules for each group pair, better predictions could be achieved, which will be part our future work. Nonetheless, if dilute gas viscosity is calculated in the same way, the RES approach yields similar statistical agreement with the experimental data as the REFPROP-models, while the RES approach has much simpler formulation and fewer parameters.

Our next goal is to explore the application of our RES approach to refrigerant (group 3) and lubricant (might be group 6) mixtures. Modeling viscosity of asymmetric mixtures remains a major challenge (Thol & Richter 2021) nowadays, and overcoming this challenge is one of the main goals in subproject 3 of the KETEC project. Modeling thermophysical properties of lubricants is demanding itself as commercially available lubricants are all mixtures with components and compositions that are difficult to determine accurately. Traditional thermophysical modelling approaches cannot be used for such lubricants, because these models are generally developed for pure fluids or mixtures with known components and compositions. A modelling approach for lubricants has been proposed by our research group (Yang et al. 2023) that assumes each lubricant to be a pseudo-pure fluid, characterizes each lubricant by fluid constants (e.g., critical temperature), and determines these fluid constants with a minimal number of experiments. This modelling approach will be combined with our current RES method for the study of viscosity of refrigerant and lubricant mixtures.

ACKNOWLEDGEMENTS

This work is funded by the German Federal Ministry of Education and Research under grant number 03SF0623A/B/C on the basis of a resolution of the German Bundestag. The authors also thank the Project Management Organization Jülich for supporting the project. The responsibility for the content of this publication lies with the authors.

NOMENCLATURE

р	pressure (Pa)	R	molar gas constant (8.3144 J mol ⁻¹ K ⁻¹)
Т	temperature (K)	V	molar volume (m ³ mol ⁻¹)
η	viscosity (Pa s)	S	molar entropy (J mol ⁻¹ K ⁻¹)
т	mass (kg)	$k_{\scriptscriptstyle B}$	Boltzmann constant (1.380649·10 ⁻²³ J K ⁻¹)
σ	collision diameter of L-J particle (nm)	Ω ^{(2,2)*}	reduced collision integral
ε/k _B	reduced L-J pair-potential energy	$ ho_{ m N}$	number density (m ⁻³)
n _k	fluid-specific fitted parameter	ξ	fluid-specific scaling factor
Xi	mole fraction of component <i>i</i>	T^*	$k_{\rm B}T/\varepsilon$ is the dimensionless temperature

REFERENCES

- Al Ghafri S.Z.S., Akhfash M., Hughes T.J., Xiao X., Yang X., May E.F. 2021. High pressure viscosity measurements of ternary (methane + propane + heptane) mixtures. *Fuel Process. Technol.* 223, 106984
- Bell I.H. 2019. Probing the link between residual entropy and viscosity of molecular fluids and model potentials. *PNAS*. 116(10), 4070–79
- Bell I.H. 2020a. Entropy Scaling of Viscosity—I: A Case Study of Propane. J. Chem. Eng. Data. 65(6), 3203–15
- Bell I.H. 2020b. Entropy Scaling of Viscosity—II: Predictive Scheme for Normal Alkanes. J. Chem. Eng. Data. 65(11), 5606–16
- Bell I.H., Delage-Santacreu S., Hoang H., Galliero G. 2021. Dynamic Crossover in Fluids: From Hard Spheres to Molecules. J. Phys. Chem. Lett. 12(27), 6411–17
- Bell I.H., Laesecke A. 2016. Viscosity of refrigerants and other working fluids from residual entropy scaling. 16th International Refrigeration and Air Conditioning Conference at Purdue, July 11-14
- Bell I.H., Messerly R., Thol M., Costigliola L., Dyre J.C. 2019. Modified Entropy Scaling of the Transport Properties of the Lennard-Jones Fluid. *J. Phys. Chem. B.* 123(29), 6345–63
- Bell I.H., Wronski J., Quoilin S., Lemort V. 2014. Pure and Pseudo-pure Fluid Thermophysical Property Evaluation and the Open-Source Thermophysical Property Library CoolProp. Ind. Eng. Chem. Res. 53(6), 2498–2508

- Binti Mohd Taib M., Trusler J.P.M. 2020. Residual entropy model for predicting the viscosities of dense fluid mixtures. J. Chem. Phys. 152(16), 164104
- Fouad W.A. 2020. Thermal Conductivity of Pure Fluids and Multicomponent Mixtures Using Residual Entropy Scaling with PC-SAFT—Application to Refrigerant Blends. J. Chem. Eng. Data. 65(12), 5688–97
- Frenkel M., Chirico R.D., Diky V., Yan X., Dong Q., Muzny C. 2005. ThermoData Engine (TDE): Software Implementation of the Dynamic Data Evaluation Concept. J. Chem. Inf. Model. 45(4), 816–38
- Gross J., Sadowski G. 2001. Perturbed-Chain SAFT: An Equation of State Based on a Perturbation Theory for Chain Molecules. *Ind. Eng. Chem. Res.* 40(4), 1244–60
- Hirschfelder J.O., Curtiss C.F., Bird R.B., Mayer M.G. 1964. *Molecular Theory of Gases and Liquids*, Vol. 165. Wiley New York
- Hopp M., Gross J. 2017. Thermal Conductivity of Real Substances from Excess Entropy Scaling Using PCP-SAFT. Ind. Eng. Chem. Res. 56(15), 4527–38
- Hopp M., Mele J., Hellmann R., Gross J. 2019. Thermal Conductivity via Entropy Scaling: An Approach That Captures the Effect of Intramolecular Degrees of Freedom. *Ind. Eng. Chem. Res.* 58(39), 18432–38
- Kim D., Liu H., Yang X., Yang F., Morfitt J., et al. 2021. Thermal conductivity measurements and correlations of pure R1243zf and binary mixtures of R32 + R1243zf and R32 + R1234yf. *International Journal of Refrigeration*. 131, 990–99
- Lemmon E.W., Bell I.H., Huber M.L., McLinden M.O. 2018. NIST Standard Reference Database 23: Reference Fluid Thermodynamic and Transport Properties-REFPROP, Version 10.0, National Institute of Standards and Technology
- Liu H., Yang F., Yang Z., Duan Y. 2020. Modeling the viscosity of hydrofluorocarbons, hydrofluoroolefins and their binary mixtures using residual entropy scaling and cubic-plus-association equation of state. *J. Mol. Liq.* 308, 113027
- Liu H., Yang F., Yang X., Yang Z., Duan Y. 2021. Modeling the thermal conductivity of hydrofluorocarbons, hydrofluoroolefins and their binary mixtures using residual entropy scaling and cubic-plusassociation equation of state. *J. Mol. Liq.* 330, 115612
- Lötgering-Lin O., Fischer M., Hopp M., Gross J. 2018. Pure Substance and Mixture Viscosities Based on Entropy Scaling and an Analytic Equation of State. *Ind. Eng. Chem. Res.* 57(11), 4095–4114
- Neufeld P.D., Janzen A.R., Aziz R.A. 1972. Empirical Equations to Calculate 16 of the Transport Collision Integrals Ω(I, s)* for the Lennard-Jones (12–6) Potential. J. Chem. Phys. 57(3), 1100–1102
- Span R., Beckmüller R., Hielscher S., Jäger A., Mickoleit E., et al. 2020. TREND. Thermodynamic Reference and Engineering Data 5.0. Bochum, Germany: Lehrstuhl für Thermodynamik, Ruhr-Universität Bochum
- Thol M., Richter M. 2021. Dynamic Viscosity of Binary Fluid Mixtures: A Review Focusing on Asymmetric Mixtures. *Int. J. Thermophys.* 42(11), 161
- Tiesinga E., Mohr P.J., Newell D.B., Taylor B.N. 2021. CODATA Recommended Values of the Fundamental Physical Constants: 2018. J. Phys. Chem. Ref. Data. 50(3), 033105
- Wang X., Wright E., Gao N., Li Y. 2020. Evaluation on Excess Entropy Scaling Method Predicting Thermal Transport Properties of Liquid HFC/HFO Refrigerants. J. Therm. Sci. 31, 1–11
- Wilke C.R. 1950. A Viscosity Equation for Gas Mixtures. J. Chem. Phys. 18(4), 517–19
- Yang X., Hanzelmann C., Feja S., Trusler J.P.M., Richter M. 2023. Thermophysical property modelling of quasipure lubricant oils and mixtures using small amount of experiments. *Ind. Eng. Chem. Res.* 0(0), (to be submitted)
- Yang X., Kim D., May E.F., Bell I.H. 2021a. Entropy Scaling of Thermal Conductivity: Application to Refrigerants and Their Mixtures. *Ind. Eng. Chem. Res.* 60(35), 13052–70
- Yang X., Liu H., Chen S.H., Kim D., Yang F., et al. 2021b. Viscosity of binary refrigerant mixtures of R32 + R1234yf and R32 + R1243zf. *Int. J. Refrig.* 128, 197–205
- Yang X., Xiao X., May E.F., Bell I.H. 2021c. Entropy Scaling of Viscosity—III: Application to Refrigerants and Their Mixtures. J. Chem. Eng. Data. 66(3), 1385–98
- Yang X., Xiao X., Thol M., Richter M., Bell I.H. 2022. Linking Viscosity to Equations of State Using Residual Entropy Scaling Theory. *Int J Thermophys.* 43(12), 183