

Viscosity of refrigerants and other working fluids from residual entropy scaling*

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ABSTRACT

We apply a modified scaling of viscosity with residual entropy in a manner analogous to that proposed by Rosenfeld (1977). We consider the viscosity-entropy scaling for a reference fluid (refrigerant R124 (1-chloro-1,2,2,2-tetrafluoroethane)), and demonstrate that the viscosity for many other refrigerants scales with residual entropy in a similar manner. Finally, we use a corresponding-states formalism with one adjustable parameter to obtain the viscosity in a non-iterative fashion for a number of refrigerants. The viscosities of the studied fluids are represented within average absolute deviations of 10%. The evaluation of the liquid viscosity from this formulation is approximately 40 times faster than evaluating the viscosity from extended corresponding states. Finally, this scheme can be used to estimate viscosities for refrigerants with a very small subset of experimental measurements, perhaps as few as one saturated liquid viscosity data points.

1. INTRODUCTION

1.1 Current practice

The dynamic modeling of thermal systems and their components has gained an increased importance in recent years. Viscosity is one of the transport properties of fluids that are required for detailed analyses of system components, including compressors, heat exchangers, and other devices. The current state-of-the-art for the transport properties of a number of working fluids of relevance are based on extended corresponding states, which is extremely computationally expensive due to the conformal state solver. More computationally efficient models for transport properties of pure fluids with reasonable accuracy are required.

A number of different models exist for the viscosity of pure fluids. The highest accuracy viscosity representations for refrigerants are obtained from fluid-specific correlations. For example, reference viscosity correlations, which can represent all the most accurate measurements to within their experimental uncertainties, are available for refrigerants R134a (Huber et al., 2003) and R125 (Huber and Laesecke, 2006). Other less-accurate viscosity correlations are available for refrigerants R123 (Tanaka and Sotani, 1996) and R23 (Shan et al., 2000).

Aside from these fluid-specific correlations, the remaining refrigerants are primarily modeled through the use of extended corresponding states. The use of extended corresponding states is a viscosity mapping methodology developed at NIST (Ely and Hanley, 1981, 1983; Huber and Ely, 1992; Klein et al., 1997; McLinden et al., 2000) and implemented in the reference thermophysical property software NIST REFPROP 9.1 (Lemmon et al., 2013). A limited subset of extended corresponding state models is also available in the CoolProp thermophysical property library (Bell et al., 2014).

Extended corresponding states is based on mapping the η - T - ρ surface for the fluid of interest onto a fluid with an accurate equation of state as well as a high-accuracy viscosity correlation. For most refrigerants, the reference fluid is either R134a or propane. To obtain the viscosity for the fluid of interest, a so-called conformal state must be obtained for the reference fluid. This conformal state is the state point where both the reference fluid and the fluid of interest have

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the same residual Helmholtz energy α and compressibility factor Z . Only a small number of required density correction polynomials (without which extended corresponding states operates in a fully-predictive manner) are available in the literature (Huber et al., 2003; McLinden et al., 2000). Unfortunately, the algorithm required to obtain the conformal state is not adequately described in the existing literature on corresponding states, and furthermore, there are numerous possible solutions, which makes the conformal state solver algorithm very fragile. The extended corresponding states viscosity models are extremely slow because a two-dimensional solver for the conformal temperature and density is required.

1.2 Entropy scaling

In the 1970s, Rosenfeld (1977) proposed a monovariate relationship between the viscosity of fluids and the residual entropy. Rosenfeld's study was based on the analysis of the soft-sphere potential as well as a small amount of simulation data for the Lennard-Jones intermolecular potential. Rosenfeld found that for a subset of substances, the viscosity was nearly a monovariate function of the residual entropy. The residual entropy is the total entropy at a thermodynamic state minus the ideal-gas entropy evaluated at the same temperature and density.

In recent years, Novak (2011; 2013) and Lötgering-Lin and Gross (2015) applied the so-called Rosenfeld scaling theory to a wider range of real fluids. Both have demonstrated that over a wide range of states, the viscosity scales in a monovariate fashion with the residual entropy obtained from SAFT equations of state. Below we describe how to evaluate the residual entropy for the Helmholtz-energy-explicit equations of state that have become the preferred means of constructing high-accuracy equations of state.

2. METHODOLOGY

2.1 Data collection

In this work we obtained experimental viscosity data for the refrigerants from the ThermoData Engine database (TRC, 2016) maintained by the National Institute of Standards and Technology. This database includes a significant amount of experimental viscosity data, and a methodology to assess the reliability of a given experimental dataset.

We have selected a number of refrigerants of interest for their novelty (refrigerants R1234yf and R1234ze(E)) or their excellent data coverage (refrigerants R125 and R134a). Figure 1 shows the size, shape, and three-dimensional charge distributions of a few of the molecules under study, which gives a physical sense for fluids considered here, and their similarity to the reference fluid selected. A list of the selected refrigerants is provided in Table 2.

2.2 Residual Entropy

The equations of state in use in NIST REFPROP (Lemmon et al., 2013) are expressed in the form of the non-dimensionalized Helmholtz energy $\alpha = A/(RT)$ as a function of temperature and density, from which all other properties can be obtained because it is a thermodynamic potential. The non-dimensionalized Helmholtz energy α is further expressed the summation of ideal-gas α^0 and residual α^r contributions. For more information, see the book of Span (2000) or the equation of state of nitrogen from Span *et al.* (2000).

The total entropy is obtained from

$$\frac{s}{R} = \tau \left[\left(\frac{\partial \alpha^r}{\partial \tau} \right)_{\delta} + \left(\frac{\partial \alpha^0}{\partial \tau} \right)_{\delta} \right] - \alpha^r - \alpha^0 \quad (1)$$

and the residual entropy from

$$\frac{s^r}{R} = \tau \left(\frac{\partial \alpha^r}{\partial \tau} \right)_{\delta} - \alpha^r, \quad (2)$$

where the independent variables of T , ρ of the Helmholtz energy potential are non-dimensionalized as the reduced density $\delta = \rho/\rho_{\text{crit}}$ and the reciprocal reduced temperature $\tau = T_{\text{crit}}/T$, where the subscript crit denotes values at the critical point.

In this work, we have found that for refrigerants the viscosity scales far better with the product of density ρ and s^r than with s^r alone, and therefore we have introduced a reduced residual entropy scaling variable given by

$$x = \rho s^r / (\rho_{\text{crit}} s_{\text{crit}}^r). \quad (3)$$

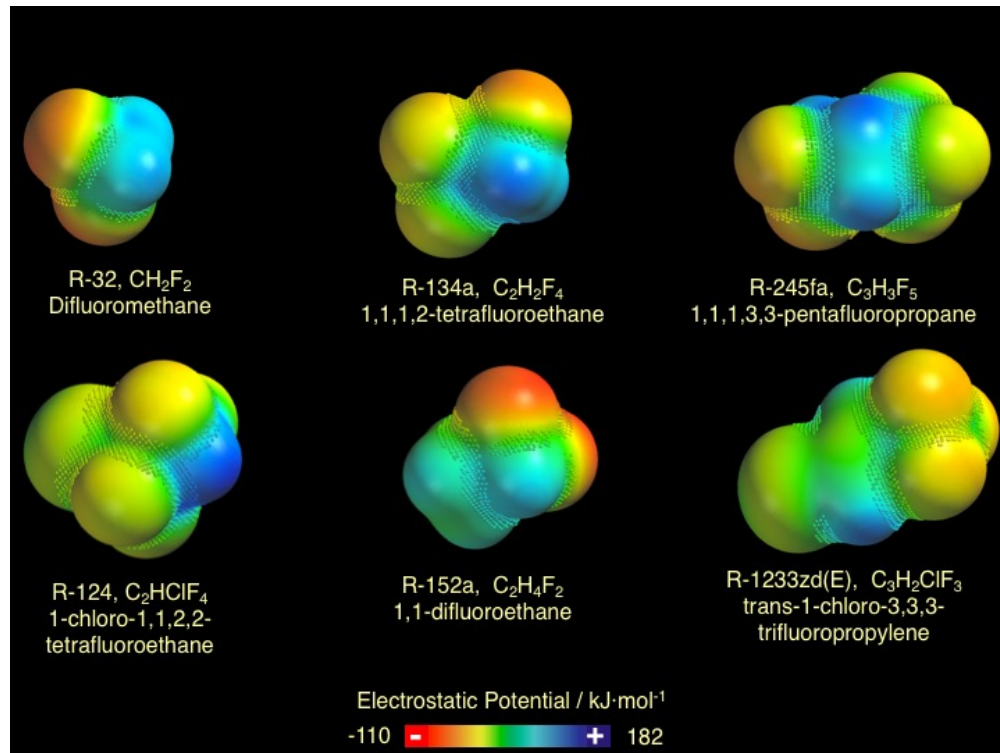


Figure 1: Molecular size, shape, and charge distribution of representative refrigerants considered in this study.

2.3 Dilute-gas viscosity

The dilute gas viscosity is based on the modified Chapman-Enskog theory (Chapman and Cowling, 1970) for monatomic gases, which yields the form

$$\eta^0 = \frac{26.692 \times 10^{-9} \sqrt{T \cdot (1000M)}}{\sigma^2 \Omega^{(2,2)}} \quad (4)$$

where the collision integral $\Omega^{(2,2)}$ is obtained from the correlation of Neufeld *et al.* (1972) for the Lennard-Jones potential

$$\Omega^{(2,2)} = 1.16145(T^*)^{-0.14874} + 0.52487 \exp(-0.77320T^*) + 2.16178 \exp(-2.43787T^*), \quad (5)$$

and the reduced temperature T^* is given by $T^* = T/(\epsilon/\kappa_B)$. All other parameters are described in the nomenclature. The parameters σ (in nm) and ϵ/κ_B (in K) are adjustable parameters related to the molecular diameter and the potential well depth, respectively.

The length and energy scaling parameters σ and ϵ/κ_B are often correlated to very low-density viscosity measurements. In this work, low-density viscosity measurements have not been made for all the fluids under study, and therefore we have decided to consistently use the estimation scheme of Chung *et al.* (1988) for all the fluids under study. In this empirical model, the length and energy scaling parameters can be estimated by the following equations:

$$\sigma = \frac{8.09}{(\rho_{\text{crit}})^{1/3}} \quad \text{and} \quad \frac{\epsilon}{\kappa_B} = \frac{T_{\text{crit}}}{1.2593}, \quad (6)$$

where the critical density ρ_{crit} is in $\text{mol}\cdot\text{m}^{-3}$, the critical temperature T_{crit} is in K, and σ is in nm.

2.4 Correlation for the reference fluid R124

In the method presented here, a reference fluid is selected, for which both a high-accuracy equation of state and a significant body of reliable experimental data are available. Unfortunately there are few refrigerants that meet both

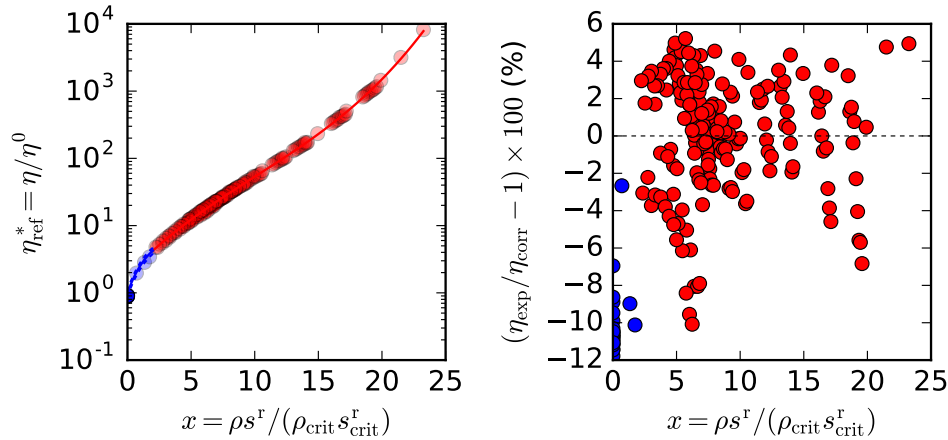


Figure 2: Scaled experimental data for refrigerant R124, correlation and deviation plot

conditions. Two of the best studied refrigerants are refrigerant R134a (1,1,1,2-tetrafluoroethane) and refrigerant R125 (1,1,1,2,2-pentafluoroethane).

Selecting refrigerant R134a as the reference fluid would be appealing due to its large body of high-accuracy experimental data (as described in a forthcoming paper (Bell et al., 2016)) and a fluid-specific viscosity formulation (Huber et al., 2003). On the other hand, the triple point temperature of refrigerant R134a is relatively high (169 K) as compared with other refrigerants like R22 (116 K) or R124 (120 K). Refrigerant R124 has one of the longest saturation curves, which makes it a suitable reference fluid for a corresponding states model.

Therefore, refrigerant R124 was selected as the reference fluid in this study, even though it has not found wide use as a refrigerant. It has experimental viscosity measurements for the saturated liquid at temperatures as low as 120 K, which allows for wide coverage of the domain of the scaled residual entropy. As seen in Figure 2, the monovariate scaling curve has two domains with different curvature and shape, roughly subdivided into the liquid region for $x > 2$ and the gaseous region for $x < 2$.

An empirical approach for modeling the viscosity of the reference refrigerant was constructed in the form

$$\ln \eta_{\text{ref}}^* = \psi_{\text{liq}} \cdot f_{\text{liq}}(x) + (1 - \psi_{\text{liq}}) \cdot f_{\text{vap}}(x) \quad (7)$$

where $\eta_{\text{ref}}^* = \eta/\eta^0$ is the reduced viscosity for the reference fluid, f_{liq} and f_{vap} are functions to model the reduced viscosity of the liquid and vapor, as discussed below, and ψ_{liq} is a Heaviside step function approximation used for the crossover

$$\psi_{\text{liq}} = \frac{1}{1 + \exp[-k(x-2)]} \quad (8)$$

where k is an adjustable parameter controlling the sharpness of the transition between liquid and vapor values. A large positive number for k results in a sharp transition; 100 works well in this case. For $x > 2$, $\psi_{\text{liq}} \approx 1$, and for $x < 2$, $\psi_{\text{liq}} \approx 0$.

The functions f_{liq} and f_{vap} are each cubic polynomials in x :

$$f_{\text{liq}} = \sum_{i=0}^3 c_{\text{liq},i} x^i \text{ and } f_{\text{vap}} = \sum_{i=0}^3 c_{\text{vap},i} x^i, \quad (9)$$

where the coefficients $c_{\text{liq},i}$ and $c_{\text{vap},i}$ are given in Table 1. The deviations between the correlation and the experimental data points are shown in Fig. 2. All experimental data points for refrigerant R124 are represented within 12%. The liquid function is obtained by fitting all the reduced viscosity data for refrigerant R124 for $x > 2$. The vapor function f_{vap} is constrained to have the same slope and derivative as f_{liq} at $x = 2$, to have a value of $\ln \eta^* = 0$ ($\eta^* = 1$) at $x = 0$, and the derivative value $\left. \frac{d(\ln \eta^*)}{dx} \right|_{x=0} = 1.2$, which was selected by manual optimization.

Table 1: Coefficients for the reference function for refrigerant R124

i	$c_{\text{liq},i}$	$c_{\text{vap},i}$
0	0.6100913843	0.0
1	0.4508958312	1.2
2	-0.017063	-0.308598
3	0.000564	0.035317

2.5 Correlation for fluid of interest

For each fluid under study, the reduced viscosity is expressed in the form

$$\eta_{\text{fluid}}^*(x) = 1 + C(\eta_{\text{ref}}^*(x) - 1) \quad (10)$$

where C is a fluid-specific constant used to scale the correlation for η_{ref}^* to the fluid of interest. In practice, rather than minimizing the error between η_{fluid}^* from Eq. 10 and the values of η^* from experiments, we minimize the error between the actual measured viscosity η and that from Eq. 10 multiplied by the dilute-gas viscosity.

To calculate the value for η for the fluid of interest, it would simply be required to evaluate $x = \rho s^t / (\rho_{\text{crit}} s_{\text{crit}}^t)$ for a state point of the fluid of interest, evaluate Eq. 7 at the calculated value of x to get η_{ref}^* , and use Eq. 10 to evaluate η^* for the fluid of interest. Finally, it is necessary to evaluate the dilute-gas viscosity η^0 from Eqs. 4 to 6 for the fluid of interest, and obtain

$$\eta_{\text{fluid}} = \eta_{\text{fluid}}^* \eta_{\text{fluid}}^0 \quad (11)$$

Values for the constant C obtained from a bounded optimization (through the use of the `fminbound` function from the `scipy.optimize` python module (Jones et al., 2001)) are given in Table 2.

Table 2: Information on the fluids of interest (based on refrigerant R124 as the reference fluid)

Fluid	AAD (%)	C	Equation of state reference
R22	15.21	0.66820	Kamei et al. (1995)
R32	4.30	0.79022	Tillner-Roth and Yokozeki (1997)
R152a	6.82	0.86166	Outcalt and McLinden (1996)
R1234yf	3.74	0.87302	Richter et al. (2011)
R1234ze(E)	4.25	0.91012	Thol and Lemmon (2016)
R124	3.58	0.99792	de Vries et al. (1995)
R134a	5.30	1.01318	Tillner-Roth and Baehr (1994)
R125	3.85	1.05714	Lemmon and Jacobsen (2005)
R245fa	7.40	1.21398	Akasaka et al. (2015)
R1233zd(E)	2.22	1.24740	Mondejár et al. (2015)

An example is provided here as a demonstration for saturated liquid R32 at 273.15 K. More digits than can be justified by uncertainty are provided for ease of validation:

- T : 273.15 K
- ρ : 20284.0588661 mol·m⁻³
- x : 5.73526520861
- M : 0.052024 kg/mol
- σ : 0.402001631822 nm
- ϵ/κ_B : 278.928769952 K
- $\Omega^{(2,2)}$: 1.60983558487
- η_{fluid}^0 : 1.22305625771 × 10⁻⁵ Pa·s
- ψ_{liq} : 1.0
- η_{ref}^* : 15.5061444226
- η_{fluid}^* : 12.4630454456
- η : 0.000152430057224 Pa·s

The value for the viscosity of saturated liquid R32 at 273.15 K from NIST REFPROP is equal to 0.0001505 Pa·s, or a difference of 1.3%.

3. RESULTS

Figure 3 shows the scaled results for each of the fluids studied in this work, applying the optimal scaling parameter C from Table 2. In this figure, the optimal values for C are *not* found when the scaled data for η_{fluid}^* are co-incident with the curve for η_{ref}^* . This offset is caused by the fact that we have reduced the experimental viscosity data by the dilute-gas viscosity values obtained from the Chung model (as described above), which introduces a significant error in the prediction of the dilute-gas viscosity. This error in the Chung model tends to be roughly constant over the entire temperature range, which results in a constant logarithmic offset. Nonetheless, the essential point to note is that the *shape* of the curves are all similar to those of the reference fluid. Therefore, the error in the dilute-gas predictive scheme is mostly cancelled out for liquid viscosities; vapor viscosities are represented with higher deviations up to 20 %, and their representation could be improved with dedicated dilute-gas equations.

Figure 4 shows the errors in the predictions of viscosity from the scaling approach used in this work. The diagrams are thresholded to the range $\pm 20\%$; values outside this range are plotted as having the limiting value. Refrigerant R124 serves as the reference fluid, which is advantageous here because this fluid has a long vapor pressure curve. Aside from refrigerant R22, which shows pronounced deviations significantly greater than 20% at low temperatures (high x), this scaling approach predicts more than 95% of the liquid viscosities within $\pm 10\%$. Deviations are greater in the vapor phase, but are still generally within $\pm 20\%$. Optimization of the Lennard-Jones potential variables could yield even more accurate predictions in the vapor phase.

3.1 Speed comparison

As computational efficiency is one of the primary motivations for this work, we present a small benchmark study to evaluate the computational speed of this method as compared with extended corresponding states. In this case, we compare the computational speed required for a large number of evaluations of the saturated liquid viscosity (between 273.15 K and 274.15 K) for refrigerant R32 from REFPROP. A modified version of REFPROP was employed that allows for the evaluation of the viscosity from the extended corresponding states model without also evaluating the thermal conductivity. Furthermore, we present the computational efficiency results for R125, for which a fluid-specific viscosity correlation was developed (Huber and Laesecke, 2006).

When compiling the sample code in Microsoft Visual Studio 2012 with the Intel Fortran compiler, the following results are obtained:

Table 3: Sample computational efficiency results

Fluid	η from REFPROP ($\mu\text{s}/\text{call}$)	This work ($\mu\text{s}/\text{call}$)	Speedup
R32	6.1	0.15	42x
R125	1.7	0.14	12x

As a point of reference, on this machine, the evaluation of one call of $p = f(T, \rho)$ without any phase determination takes approximately $0.9 \mu\text{s}$.

CONCLUSIONS

The following conclusions can be obtained from this study:

- A seldomly considered relation between the thermodynamic property entropy and the transport property viscosity was applied over the entire fluid region in a single-parameter corresponding states framework for ten refrigerants.
- This framework provides a relation between these two properties which should be developed further.
- Of the refrigerants under study, six are currently modeled through the use of the extended corresponding states method in state-of-the-art libraries of thermophysical properties.
- The average absolute deviations in the current formulations ranged from from 2.2% to 7.4%, except for R22, which had an average absolute deviation of 15.2%.
- The computational speed of this model is approximately 40 times faster than extended corresponding states.

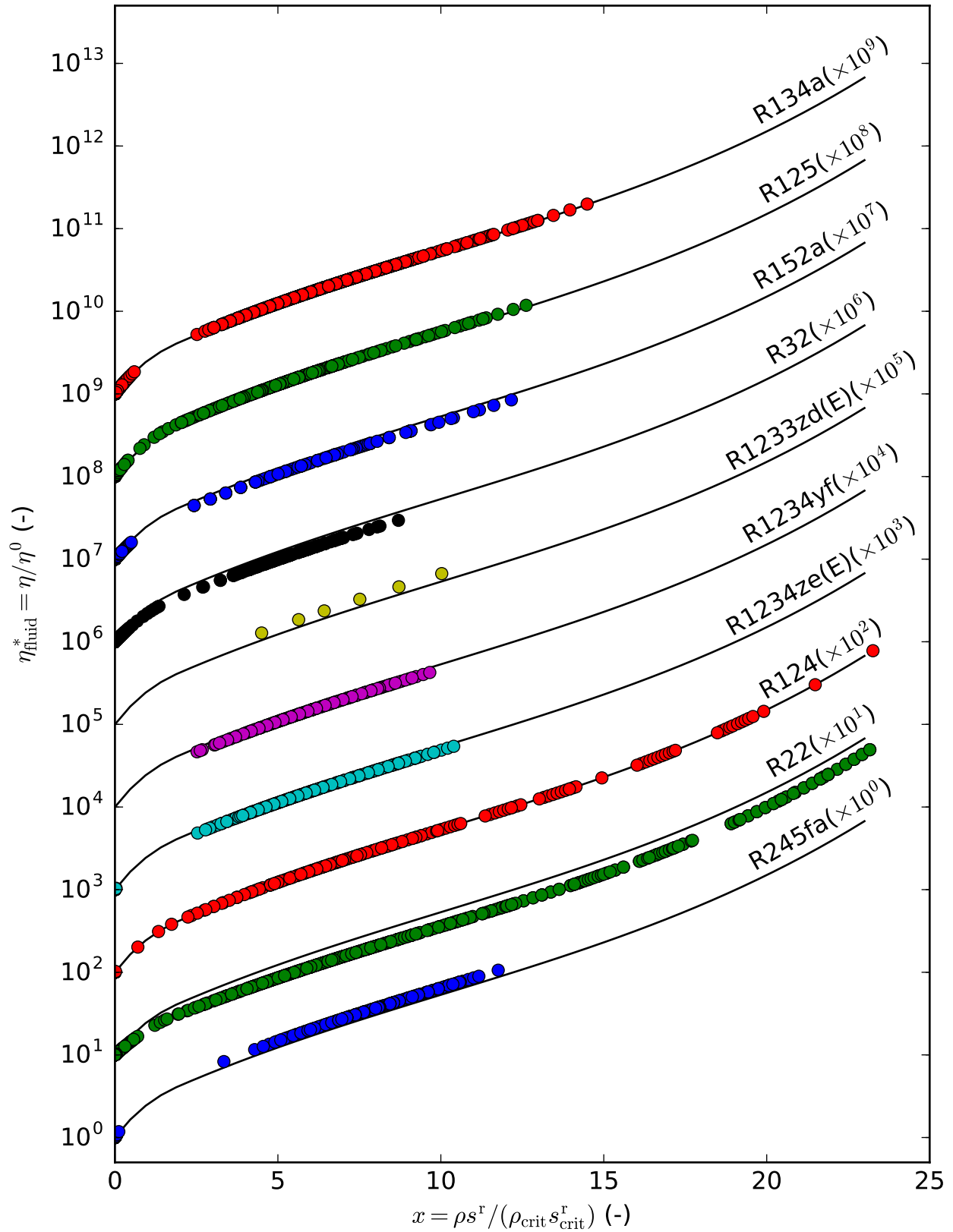


Figure 3: Scaled viscosity data for working fluids under study (solid line: refrigerant R124 reference function)

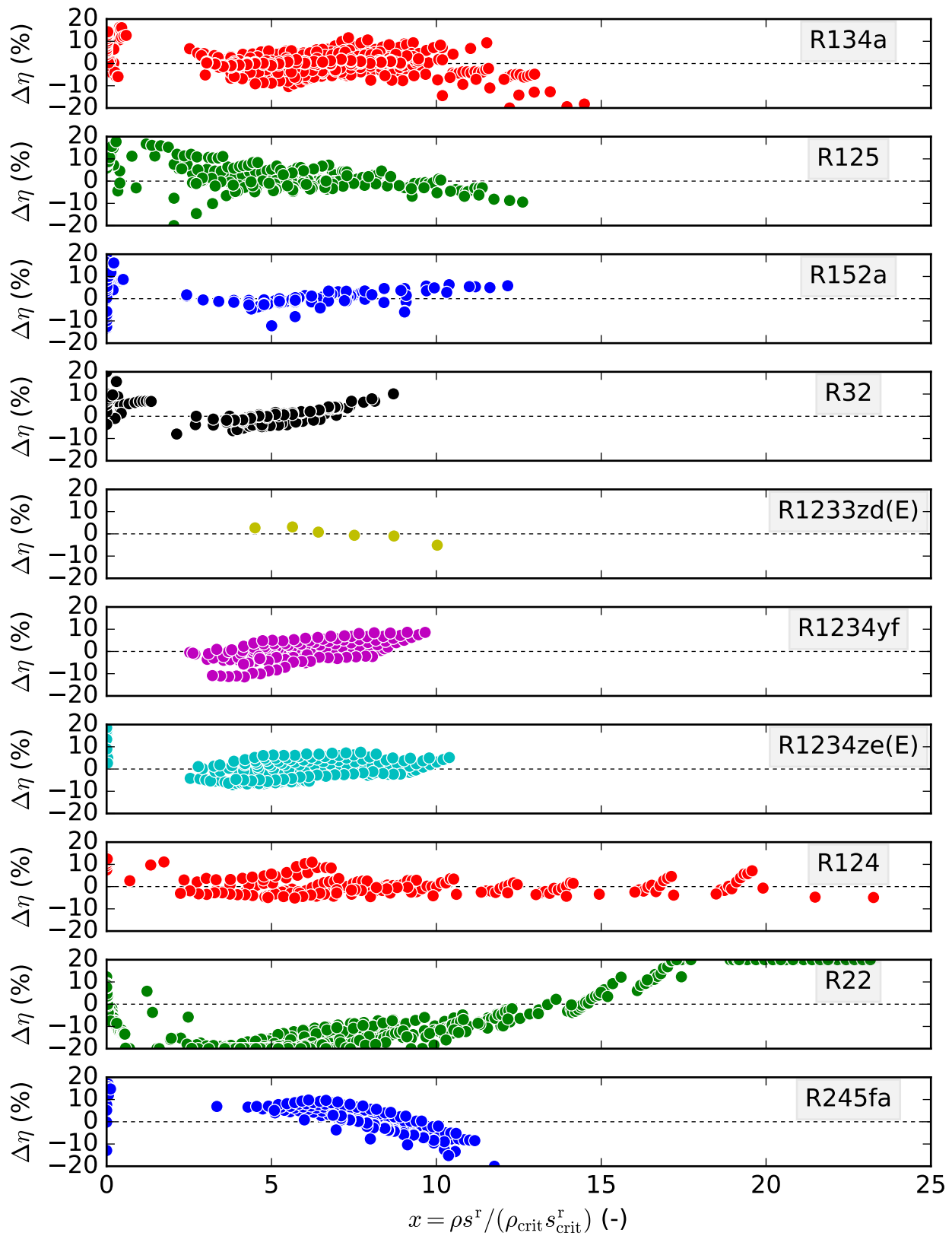


Figure 4: Error in prediction of viscosity for the working fluids under study (error data thresholded to $\pm 20\%$)

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NOMENCLATURE

A	Helmholtz energy ($\text{J}\cdot\text{mol}^{-1}$)	δ	Reduced density (-)
$c_{\text{liq},i}$	Constants for liquid correlation	a	Helmholtz free energy (-)
$c_{\text{vap},i}$	Constants for vapor correlation	a^r	Residual Helmholtz free energy (-)
C	Viscosity scaling factor (-)	a^0	Ideal-gas Helmholtz free energy (-)
k	Crossover factor (-)	ϵ/κ_B	Energy parameter for Lennard-Jones (K)
M	Molar mass ($\text{kg}\cdot\text{mol}^{-1}$)	η	Viscosity ($\text{Pa}\cdot\text{s}$)
f_{liq}	Correlation function for liquid	η^0	Dilute-gas viscosity ($\text{Pa}\cdot\text{s}$)
f_{vap}	Correlation function for vapor	η_{ref}^*	Reduced viscosity for reference fluid (-)
s	Entropy ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)	η_{fluid}^*	Reduced viscosity for fluid of interest (-)
s^r	Residual entropy ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)	ψ_{liq}	Crossover variable (-)
s_{crit}^r	Critical residual entropy ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)	$\Omega^{(2,2)}$	Collision integral (-)
R	Universal gas constant ($\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$)	ρ	Molar density ($\text{mol}\cdot\text{m}^{-3}$)
T	Temperature (K)	ρ_{crit}	Critical molar density ($\text{mol}\cdot\text{m}^{-3}$)
T_{crit}	Critical temperature (K)	σ	Size parameter for Lennard-Jones (nm)
T^*	Energy reduced temperature (-)	τ	Reciprocal reduced temperature (-)

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