

A Reference Correlation for the Viscosity of Krypton From Entropy Scaling

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Abstract

We present a new wide-ranging correlation for the viscosity of krypton based on critically evaluated experimental data. For the first time, such a correlation has as its basis the entropy scaling approach. We base the residual contribution on the Lennard-Jones fluid, resulting in one adjustable parameter for the entire phase diagram away from the dilute-gas limit. The estimated uncertainty is less than 2.0% (at the 95% confidence level) over the entire phase diagram, except in the extended critical region. The correlation is valid from 70 K to 5000 K for the dilute gas, and from 115.775 K to 750 K in the fluid phase, with a pressure limit equal to that of the melting curve.

1 Introduction

In a series of recent papers, reference correlations for the viscosity of selected common fluids [1–8] have been developed that cover a wide range of temperature and pressure conditions, including the gas, liquid, and supercritical phases. In this paper the residual entropy scaling approach is used to generate a reference viscosity correlation for krypton.

1.1 Background

The currently employed reference correlation for the viscosity of krypton was developed by Hanley et al. [9] in 1974; it is based on the corresponding-states principle

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and covers a temperature range (125 to 500) K and pressures up to 20 MPa. The only other available correlation is the corresponding-states model developed by Huber [10] and implemented in REFPROP v10.0 [11]; the documentation of REFPROP 10 states for this model: "Uncertainty of viscosity in the liquid phase is 30%, data unavailable. Uncertainty of viscosity in the gas phase at atmospheric pressure is 3%"

The analysis that will be described is based upon the best available experimental data for the viscosity. Thus, a prerequisite to the analysis is a critical assessment of the experimental data. For this purpose, two categories of experimental data are defined: primary data, employed in the development of the correlation, and secondary data, used simply for comparison purposes. According to the recommendation adopted by the Subcommittee on Transport Properties (now known as the International Association for Transport Properties) of the International Union of Pure and Applied Chemistry, the primary data are identified by a well-established set of criteria [12]. These criteria have been successfully employed to establish standard reference values for the viscosity and thermal conductivity of fluids over wide ranges of conditions, with uncertainties in the range of 1%. However, in many cases, such a narrow definition unacceptably limits the range of the data representation. Consequently, within the primary data set, it is also necessary to include results that extend over a wide range of conditions, albeit with a higher uncertainty, provided they are consistent with other lower uncertainty data or with theory. In all cases, the uncertainty claimed for the final recommended data must reflect the estimated uncertainty in the primary information.

1.2 Formulation

Historically, empirical viscosity correlations have been based upon a formulation similar to

$$\eta(T, \rho) = \eta_{\rho \rightarrow 0}(T) + \eta_1(T, \rho) + \eta^{\text{res}}(T, \rho) + \eta^{\text{crit}} \quad (1)$$

in which T is the temperature, ρ is the molar density, $\eta_{\rho \rightarrow 0}$ is the viscosity contribution in the dilute-gas limit, which can in some cases be obtained by first principles calculations or low density measurements extrapolated to zero density. The residual contribution η^{res} accounts for the collective viscosity arising from many-body interactions. The initial density contribution η_1 (which is linear in density [13, 14]) and the critical enhancement contribution η^{crit} are not considered in this work. There is insufficient data in the critical region to fit the critical enhancement term and the initial density term does not appear to be necessary for krypton (see discussion below).

Recent works relating to entropy scaling [15–18] highlight that the residual entropy is an independent variable which captures most of the dependence of the appropriately scaled transport properties, such that the scaled version of $\eta^{\text{res}}(T, \rho)$ becomes instead a quasi-monovariate function of the residual entropy. The residual entropy is defined by

$$s^{\text{r}} \equiv s(T, \rho) - s^{\text{ig}}(T, \rho) \quad (2)$$

where s is the overall entropy, and s^{ig} is the ideal-gas entropy. The residual portion of an equation of state (EOS), such as those in use in REFPROP [19] or CoolProp [20], is expressed in the form $\alpha^{\text{r}}(T, \rho)$. The quantity $\alpha^{\text{r}} = a^{\text{r}}/(RT)$ is the

reduced residual Helmholtz energy, with a^r the residual Helmholtz energy, such that α^r is non-dimensional. Thus the residual entropy is in practice evaluated from derivatives of the residual Helmholtz energy

$$\frac{s^r}{R} = -T \left(\frac{\partial \alpha^r}{\partial T} \right)_\rho - \alpha^r \quad (3)$$

Here s^r is the molar residual entropy and R is the universal gas constant used in the equation of state.

In order to simplify the discussion to follow, the quantity s^+ is defined by

$$s^+ \equiv -\frac{s^r}{R} \quad (4)$$

The entropy scaling approach for viscosity [15, 16, 18, 21, 22], which was first proposed by Rosenfeld [23, 24] suggests an alternative formulation in which the shear viscosity is first macroscopically scaled (divided by the mass, length, and energy dimensions in the liquid phase), defining the macroscopically scaled viscosity

$$\tilde{\eta} = \frac{\eta}{\rho_N^{2/3} \sqrt{m k_B T}} \quad (5)$$

in which η is the shear viscosity, ρ_N is the number density of atoms per volume, m is the mass of one atom, k_B is Boltzmann's constant, and T is the temperature. Base-SI units are used consistently (kg, m, s, Pa, etc.); the quantity $\tilde{\eta}$ is non-dimensional.

The quantity $\tilde{\eta}$ diverges in the limit of zero number density and a new variable η^+ is defined [17, 25, 26] to repair the divergence at zero density

$$\eta^+ \equiv \tilde{\eta} \times (s^+)^{2/3} \quad (6)$$

by analogy with the inverse-power-law fluid in the dilute-gas limit [24, 27]. A parallel formulation to Eq. 1 in plus-scaled coordinates reads

$$\eta^+ = \eta_{\rho \rightarrow 0}^+(T) + \eta^{\text{res}}(s^+) \quad (7)$$

This formula in Eq. (7) will be used in this work to build a correlation for the viscosity of krypton. By shifting to this plus-scaled formulation of entropy scaling, similarities between the residual contribution for fluids can be identified, which underpins a growing body of work on entropy scaling applied to the transport properties of molecular species. In particular, we will take advantage of the fact that $\eta^{\text{res}}(s^+)$ for krypton is nearly identical to that of the Lennard-Jones 12-6 fluid [25]; the L-J 12-6 fluid is heavily used in many estimation methods for transport properties. A novel feature of this study is that the length and energy scales of the Lennard-Jones potential are not explicitly needed, as they are “hiding” in the scaled quantities η^+ and s^+ .

This approach shares a common conceptual basis with the extended corresponding states approach [10, 28], although the entropy scaling application does not require to solve for the conformal state, which has a computational speed benefit [29].

The entropy scaling approach is connected with the isomorph theory [30–34], which is an observation from molecular dynamics studies of simple fluids that

when the Pearson correlation between the time histories of the virial energy and the potential energy of the ensemble of particles in the canonical ensemble (R_{Ros}) is greater than 0.9, certain quantities are invariant along curves of constant reduced structure. Two variables that are constant along such curves are $\tilde{\eta}$ and s^+ . The value of R_{Ros} tends to decrease on approaching the critical point and heading towards the gaseous limit (see Fig. 4 from Ref. [25]). A persistent mystery, which works in our favor in this case, is why the relationship between $\tilde{\eta}$ and s^+ is sometimes still strong, even when $R_{\text{Ros}} \ll 0.9$. In other words, it appears that $R_{\text{Ros}} > 0.9$ is a sufficient, but not necessary, condition to apply entropy scaling. Real fluids (mostly) follow entropy scaling.

2 Experimental Data

Table 1 summarizes, to the best of our knowledge, the theoretical prediction/estimations as well as the experimental measurements of the viscosity of krypton reported in the literature. Uncertainties given in Table 1 are those provided by the original authors. As early as 1962, based on kinetic-theory calculations, Svehla [35] proposed dilute-gas viscosity values covering the temperature range (100 to 5000) K. However, the first empirical correlation for the viscosity and thermal conductivity of krypton based on the corresponding-states principle was proposed in 1974 by Hanley et al. [9]. The viscosity correlation covered a temperature range from the triple point to 500 K and pressures up to 20 MPa with an uncertainty of 5%.

In 1983, Najafi et al. [36] employed an improved two-parameter corresponding-states scheme to correlate the dilute-gas viscosity from (100 to 2000) K with an uncertainty of better than 2%, while Vargaftik and Vasilevskaya [37] proposed dilute-gas viscosity values based on kinetic-theory calculations, covering a temperature range (800 to 5000) K with an uncertainty of up to 5%. In 1989 Golovicher et al. [38] proposed recommended values for krypton with 1.5% uncertainty over the temperature range (119 to 2000) K at 0.1 MPa, based on very accurate measurements of argon and corresponding states.

In 1990, Bich et al. [39] proposed reference values for the viscosity of krypton from the triple point to 5000 K at zero density and also at 0.101325 MPa, with an uncertainty ranging from 0.3% to 2% at the highest temperatures. A critical assessment of 18 viscometers, carried out by Berg and Moldover [40] in 2012, resulted in the proposition of the viscosity value of $(25.3062 \pm 0.0080) \mu\text{Pa s}$ for krypton at 298.15 K and zero density.

In 2016, Vogel [41] published low-uncertainty values of the viscosity of krypton at zero density over the temperature range (295 to 650) K. These were based on a reevaluation of their oscillating-disk measurements [42], employing a more recent value of the viscosity of argon for the calibration of the instrument. Their work was expanded by Jäger et al. [43], who produced reference values for the dilute gas over a temperature range (100 to 5000) K with an uncertainty of 0.07 to 0.28%, based on an ab initio intermolecular potential energy and related spectroscopic and thermophysical properties data for krypton.

In 2020, the combined use of experimental viscosity ratios together with ab initio calculations for helium has driven significant improvements in the description of dilute gas transport properties [44]. Hence, Xiao et al. [44, 45] first used improvements to ab initio helium calculations [46] to update previously measured

viscosity ratios [47]. Subsequently, they used these improved values to get better reference correlations for the dilute-gas viscosity of krypton and 9 other gases. The new reference dilute-gas viscosity correlation for krypton covers the temperature range from (70 to 5000) K with a relative uncertainty of 0.1%, and it will form the dilute-gas viscosity contribution of krypton in this work. We note that the uncertainties in Xiao et al. [44, 45] are expressed as standard uncertainties corresponding to a coverage factor of $k = 1$; in this work all uncertainties discussed are combined expanded uncertainties with a coverage of $k = 2$ (aside from the as-reported uncertainties in Table 1).

2.1 Data Assessment

In Table 1, measurements of the viscosity of krypton are separated into primary and secondary data. We kept all measurements with an author-reported uncertainty of up to 2% in the primary data set, while the rest were included in the secondary data set. The only exception is the measurements of Reynes and Thodos [65] which although they do not quote uncertainty, were included in the primary data set as they extend to high pressures. Furthermore, we note that we did not include the measurements of Kestin and Leidenfrost [69] in the primary data set, as they have been superseded by newer measurements of the same group [55, 58, 60].

The development of the correlation requires densities; Lemmon and Span [70] developed an accurate, wide-ranging equation of state that is valid from the triple point up to 750 K and 200 MPa. The uncertainties in density are typically 0.2% below 100 MPa, increasing to 1% at pressures up to 200 MPa. The uncertainty in vapor pressure is 0.2%, and the uncertainty in speed of sound is 1% below 20 MPa in the liquid phase, and 3% below 100 MPa at other state points. We adopt the values for the critical point from their equation of state; the critical temperature, T_c , and the critical density, ρ_c , are 209.48 K and 909.21 kg m⁻³, respectively. We also adopt the value they used for the triple-point temperature of 115.78 K.

Table 1 Theoretical predictions and measurements of the viscosity of krypton.

Authors / Ref.	Year	Technique ^a	Purity ^b %	Uncert. ^c %	N ^d	T ^e K	p MPa
Reference Correlations/Values							
Xiao et al. [44]	2020	$\eta_{\rho \rightarrow 0}$ Cor.	-	0.1	-	70-5000	0
Jäger et al. [48]	2016	Ab initio	-	0.07-0.28	109	100-5000	0
Vogel [41]	2016	RV	-	0.1-0.2	252	296-689	0
Berg and Moldover [40]	2012	RV	-	0.03	1	298.15	0
Bich et al. [39]	1990	RV	-	0.3-2.0	92	120-5000	0
			-	0.3-3.0	92	120-5000	0.1
Golovicher et al. [38]	1989	RV CS	-	1.5	33	119-2000	0.1
Vargaftik and Vasilevskay [37]	1984	KT Calc.	-	3-5	34	800-5000	0.1
Najafi et al. [36]	1983	CS Cor.	-	2	-	100-2000	0
Hanley et al. [9]	1974	CS Cor.	-	5	-	125-500	20
Svehla [35]	1962	KT Calc.	-	na	50	100-5000	0.1
Primary Data							
Humberg and Richter [49]	2019	RotCyl	99.998	0.17-0.29	46	253-473	0.1-2.6
Lin et al. [50]	2016	2Cap	99.999	0.1	17	243-393	0
Evers et al. [51]	2002	RotCyl	99.997	0.15	22	298-348	0.1-8.5
Wilhelm and Vogel [52]	2000	VBW	99.998	0.2	107	298,348	0.05-16
Vogel [42]	1984	OD	na	0.1-0.2	20	296-649	0.1
Abachi et al. [53]	1980	VBW	na	2.0	4	116-120	0.07-0.1
Ulybin et al. [54]	1978	Cap	99.975	1.4	37	130-300	8.7-41
Kestin et al. [55]	1978	OD	99.995	0.1-0.3	9	298-778	0.1
van den Berg and Trappeniers [56]	1978	Transp	na	0.01	34	298	0.1-6.8
Gough et al. [57]	1976	Cap	99.8	1.0-1.7	11	120-320	0.1
Kestin et al. [58]	1972	OD	99.994	0.1	8	298-973	0
Dawe and Smith [59]	1970	Cap	99.8	0.4	15	293-1600	0.1
Kalelkar and Kestin [60]	1970	OD	99.994	0.1	10	298-1151	0
Clarke and Smith [61]	1968	Cap	99.0	0.5	11	134-374	0.1
Boon et al. [62]	1967	Cap	na	1.0	10	116-123	0.07-0.13
Rigby and Smith [63]	1965	Cap	99.7	1	15	293-972	0.1
Trappeniers et al. [64]	1965	Transp	na	1.0	95	298-398	0.7-207
Reynes and Thodos [65]	1964	Cap	na	na	36	373,423,473	7-83
Thornton [66]	1960	Cap	99-100	1.0	1	291	0.09
Secondary Data							
Baharudin et al. [67]	1975	Interf	na	5.0	6	117-131	0.08-0.2
Clifton [68]	1963	OD	99.96	2.2	27	297-666	0.004-0.006
Kestin and Leidenfrost [69]	1959	OD	99.959	0.03	6	293	2

a: 2Cap: Double Capillary; Cap: Capillary; CS: Corresponding States; OD: Oscillating Disc; Interf: Interferometer; RotCyl: Rotating Cylinder; Transp: Transpiration technique; VBW: Vibrating Wire; RV: Recommended Values; KT: Kinetic Theory

b: na, not available.

c: as given by the authors; in some cases authors report “accuracy” or “uncertainty”

d: number of data

e: the triple-point temperature is 115.78 K

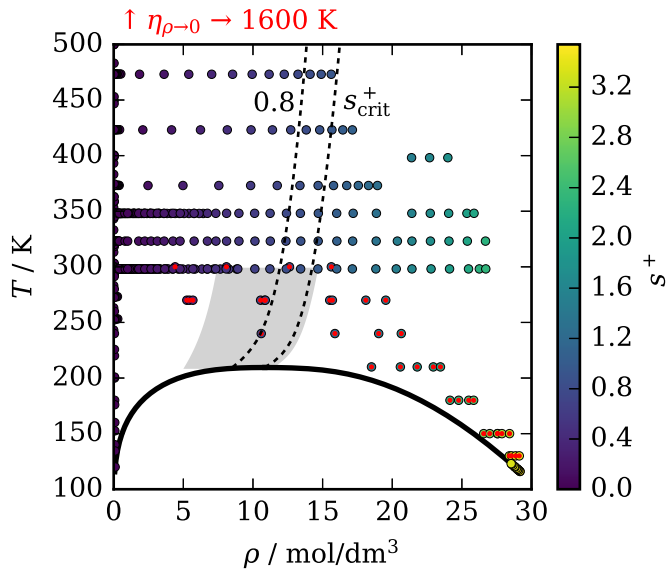


Fig. 1 Location of the data points in the temperature-density plane. Points are colored by s^+ , and the points with a red dot in the middle are from Ulybin et al. [54]. Data in the dilute gas extend to 1600 K, as indicated by the red label in the figure. The filled grey domain is the region of isomorph breakdown.

Once the densities have been obtained for each experimental data point, the data points are plotted in the temperature-density coordinates in Figure 1. The data points are colored by their value of s^+ as calculated by the equation of state. Two curves of constant s^+ are also overlaid as they will be referred to in the analysis to follow. For the temperature range greater than 300 K, the data coverage is sufficient, but for temperatures less than 298 K, the data are only present in the dilute limit and near the triple point along the saturated liquid curve, aside from one source from Ulybin et al. [54].

In light of the limited data in the extended critical region for krypton, the data for argon and the Lennard-Jones fluid are first considered in the extended critical region of the phase diagram in Figure 2. For argon, there are a few datasets telling a coherent story: there is a monovariate curve in this region of the phase diagram for “high-enough” temperatures, and the isotherm nearest the critical temperature shows at worst a deviation of approximately 5% from the higher temperature cluster of data. A similar consistent behavior is shown for the primary experimental data of propane [18]. For Lennard-Jones, the “uncertainty” in the simulation data in this region is relatively larger (estimated to be 5%), and the curve is monovariate within the uncertainty of the data. The scaling for argon and krypton should be more similar than krypton and Lennard-Jones because their pair potentials are more conformal. As a demonstration, the zero density limit of the effective hardness of interaction is nearly coincident for temperatures below the Boyle temperature for neon, argon, krypton, and xenon (see Fig. 9 of Ref. 71), a behavior not shared with Lennard-Jones. The higher the temperature, the more repulsively dominated the physics are, and the lower temperature data should fall below the bulk of the high-temperature data (at high temperatures isomorph theory becomes an increasingly good approximation). This exercise highlights how even if a fluid may not follow entropy scaling approaching the critical point, precisely how the data deviate from the monovariate behavior can highlight errors in the equation of state or the data themselves.

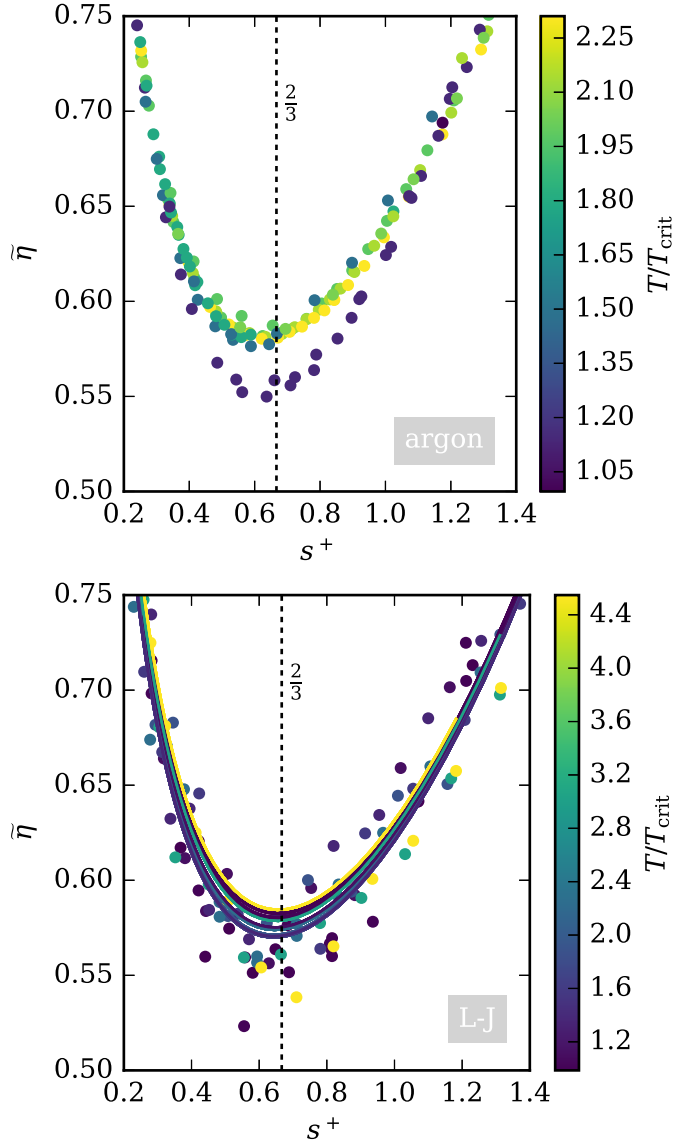


Fig. 2 Scaled experimental data of $\tilde{\eta}$ near its minima for argon [17] and Lennard-Jones [25]. The value of $s^+ = 2/3$ is included to highlight consistency with the observation [72] that the minima of $\tilde{\eta}$ occur at a value near $s^+ = 2/3$ for some monoatomic fluids.

Krypton's data in Fig. 3 do not show the expected collapse of higher-temperature ($T > 1.5T_{\text{crit}}$) datasets as in the case of argon, and it is therefore instructive to use argon's data to infer possible explanations. The reference viscosity correlations for xenon [13] and argon [73] each have mean absolute relative errors greater than 5% on data from Trappeniers, which suggests systematic errors. The high temperature data from Reynes and Thodos [65] also appear to have systematic errors

for $s^+ > 0.9$ when comparing with Figure 2. The only dataset in the temperature range from the critical temperature to room temperature away from the dilute-gas limit (see Fig. 1) is that of Ulybin et al. [54], so it is difficult to ascertain consistency with other datasets. Comparing with argon, the points around $s^+ = 0.8$ from Ulybin et al. [54] should be judged with some suspicion, and particularly the two data points with the lowest values of $\tilde{\eta}$. The dataset of Wilhelm and Vogel [52] is in good agreement with the other modern measurements in the gas phase. The ECS model implemented in REFPROP 10.0 passes through the Ulybin et al. [54] data, but does not agree with the other high-temperature data where entropy scaling should be a particularly good approach.

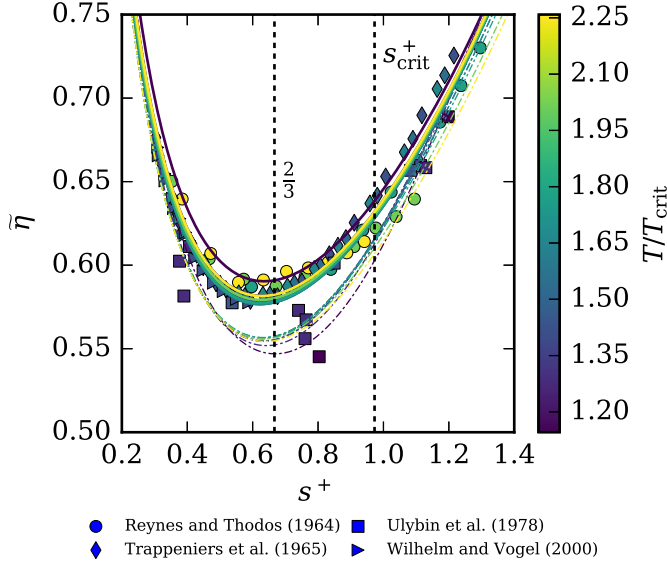


Fig. 3 Data of $\tilde{\eta}$ for krypton near their minima. Sources as listed in Table 1. Solid curves correspond to the model developed in this work, and the dashed-dotted curve is that obtained from the ECS model implemented in REFPROP 10.0[10].

3 Correlation

3.1 The Dilute-Gas Limit

The dilute-gas limit viscosity, $\eta_{\rho \rightarrow 0}(T)$, can be analyzed independently of all other contributions in Eq. (1). As already discussed in the previous section, Xiao et al. [44, 45] first used improvements to *ab initio* helium calculations [46] to update previously measured viscosity ratios [47]. Following this, they used these improved values to get better reference correlations for the dilute-gas viscosity of krypton and 9 other gases. The new reference dilute-gas viscosity correlation for krypton covers the temperature range from (70 - 5000) K with an expanded combined ($k = 2$) uncertainty of 0.1%, and it forms the dilute-gas viscosity contribution of krypton in this work. The dilute-gas viscosity, $\eta_{\rho \rightarrow 0}$ ($\mu\text{Pa}\cdot\text{s}$) given by Xiao et al. [44, 45] is,

$$\frac{\eta_{\rho \rightarrow 0}(T)}{\eta_{\rho \rightarrow 0, 298.15 \text{ K}}} = \exp \left\{ \sum_{i=1}^{12} a_i \left(\ln \left[\frac{T}{298.15 \text{ K}} \right] \right)^i \right\} \quad (8)$$

For the viscosity at room temperature $\eta_{\rho \rightarrow 0, 298.15 \text{ K}}$, the value of $(25.3062 \pm 0.0080) \mu\text{Pa}\cdot\text{s}$, proposed by Berg and Moldover, was adopted [44]. The coefficients a_i are shown in Table 2.

Table 2 Coefficients a_i of Eq. (8) [44].

i	a_i	i	a_i
1	$9.129\,712 \times 10^{-1}$	7	$2.237\,544 \times 10^{-3}$
2	$-1.001\,470 \times 10^{-1}$	8	$-1.455\,422 \times 10^{-4}$
3	$-2.454\,742 \times 10^{-2}$	9	$-2.006\,385 \times 10^{-4}$
4	$3.145\,009 \times 10^{-2}$	10	$8.341\,288 \times 10^{-5}$
5	$-4.456\,257 \times 10^{-3}$	11	$-1.520\,236 \times 10^{-5}$
6	$-4.511\,243 \times 10^{-3}$	12	$1.159\,085 \times 10^{-6}$

In the modified residual entropy scaling framework, the dilute gas term becomes [25, 27]

$$\eta_{\rho \rightarrow 0}^+ = \frac{\eta_{\rho \rightarrow 0}(T)}{\sqrt{mk_B T}} \Theta_2^{2/3} \quad (9)$$

where $\eta_{\rho \rightarrow 0}$ comes from Eq. (8) and Θ_2 is the effective cross section per molecule, equal to

$$\Theta_2 = B_2 + T \frac{dB_2}{dT}. \quad (10)$$

Values of the second virial coefficient B_2 , which comes either from the equation of state, or from integration of the potential of Ref. 43 with third-order quantum corrections (a description of the method and its implementation in Python are provided in the supporting information of Ref. [71]). In this work, the quantum-corrected values are only used as the reference data for comparisons, but the EOS is used for all calculations, and the EOS is used in the correlation so that it will be consistent with the values of s^+ obtained from the equation of state for dense states.

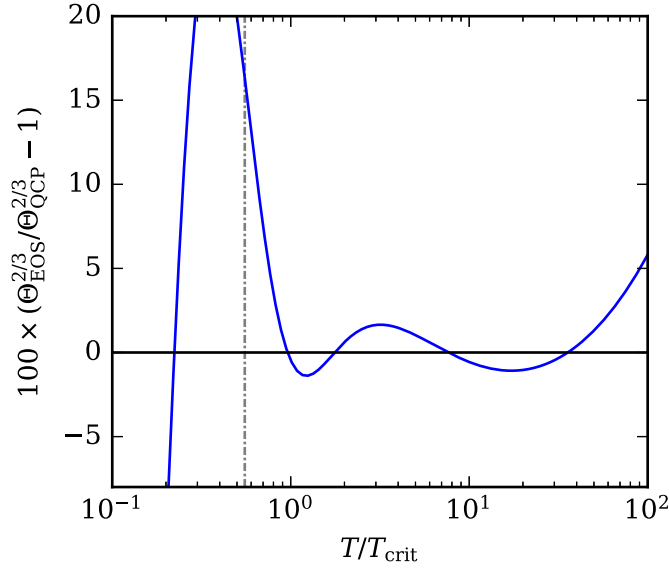


Fig. 4 Percentage deviations in $\Theta_2^{2/3}$ for krypton obtained from the second virial coefficients of the equation of state (EOS) compared with results for the pair potential with third-order quantum corrections (QCP) [27, 43]. The vertical line indicates the triple point temperature.

A new “feature” of the modified entropy scaling approach is that second virial coefficients are now invoked in order to calculate the scaled dilute gas transport properties. This poses a unique challenge as some equations of state yield clearly erroneous values for the second virial coefficients as compared with highly accurate ab initio calculations. Further detailed investigation of the adequacy of second virial coefficients for molecules where ab initio calculations are available is needed.

Fig. 4 shows the deviations between the values obtained from integration of the potential with quantum corrections and the values obtained from the EOS. Although the deviations in Fig. 4 are rather large, this poses only a minimal problem because in the dilute-gas limit this quantity is used to scale and unscale the viscosity symmetrically. On the other hand, for state points in the dense liquid near the triple point, η^+ is much larger than $\eta_{\rho \rightarrow 0}^+$ (approximately 20 times greater for krypton), and thus a relatively large error in $\eta_{\rho \rightarrow 0}^+$ comprising only a small contribution to the overall η^+ causes only a relatively modest error on the scale of the experimental uncertainty.

A further test on the residual entropy coming from the equation of state is to calculate the two-body effective hardness of interaction n_{eff} , which strongly constrains the entropy in the liquid phase, and is in general defined by derivatives at constant residual entropy[71]. In the dilute-gas limit n_{eff} is related to the second virial coefficient B_2 via:

$$n_{\text{eff}} = -3 \frac{B_2 + T \frac{dB_2}{dT}}{2T \frac{dB_2}{dT} + T^2 \frac{d^2 B_2}{dT^2}} = -3 \frac{\Theta_2}{T \frac{d\Theta_2}{dT}} \quad (11)$$

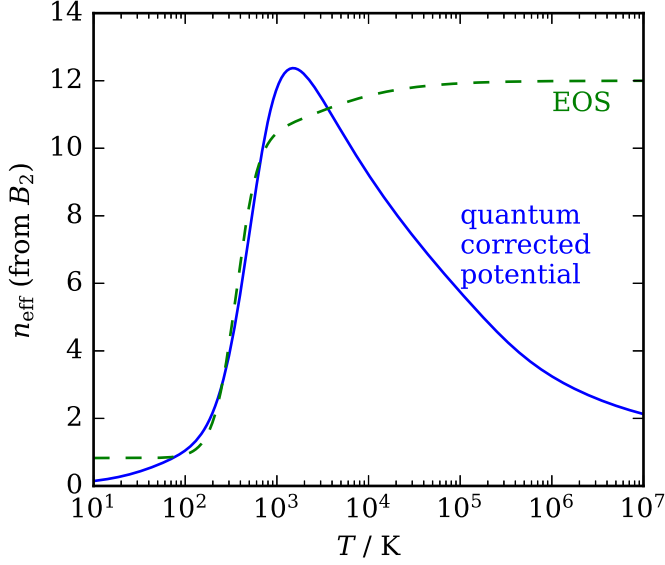


Fig. 5 Two-body effective hardness of interaction for krypton for n_{eff} from equation of state and potential with quantum corrections.

The values of n_{eff} are calculated from the EOS and compared with the values calculated from the quantum-corrected potential in Fig. 5. From this comparison it is clear that the n_{eff} from the equation of state is incorrect, both in the low-temperature limit (where the value should approach zero) and the high-temperature limit (where the value should approach $3/2$, see Section 4 for the derivation of this result). In the engineering range of temperature from approximately 100 K to 1000 K, the behavior is reasonable. It appears that the effective hardness derived from the dilute gas viscosity should also go to $3/2$ in the limit of infinite temperature, a sensitive test for high-temperature extrapolation of dilute-gas viscosity models [27].

3.2 Residual Term

Following the same approach as in other recent works, the modified residual entropy scaling approach is applied to the residual contribution. In this framework, the quantity $\eta^+ - \eta_{\rho \rightarrow 0}^+ + 1$ is expressed as a function of s^+ . This quantity is very close to a monovariate function of s^+ over much, though not all, of the phase diagram. In the case of the Lennard-Jones fluid, the collapse of the simulation data was so good that a short polynomial function in s^+ was enough to fit a reference correlation for the viscosity. We apply the same technique here in Fig. 6, overlaying the empirical curve for Lennard-Jones on the scaled data for krypton. The agreement is qualitatively excellent, perhaps surprising in that the repulsion of krypton is not perfectly represented by the Lennard-Jones potential [71].

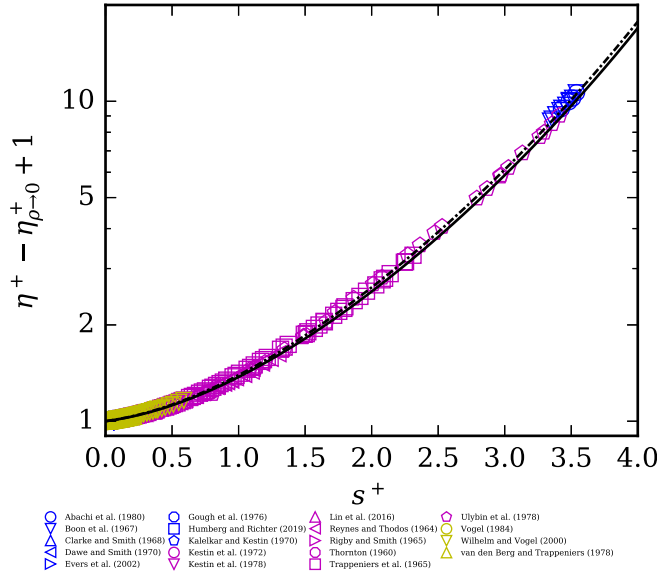


Fig. 6 Scaled residual viscosity plus 1. The solid curve is that of the empirical correlation for Lennard-Jones [25], and the dashed-dotted curve is the scaled curve. Sources as listed in Table 1.

Although the Lennard-Jones contribution for the residual portion is in excellent qualitative agreement with that of the Lennard-Jones fluid, a small empirical adjustment to the correlation is needed in order to better represent the krypton data. The residual contribution to η^+ from Lennard-Jones is multiplied by a scaling factor of 1.05, manually obtained in order to best represent the experimental data (minimizing the width of the 95% confidence interval in the liquid phase).

One might be inclined to add an initial density term to the correlation, as is commonly applied to viscosity correlations (e.g., Ref. [13, 74]), but the necessity of such a term in the entropy scaling approach is not yet clear. Figure 7 shows the plus-scaled form of the low density data, highlighting that in this case the s^+ dependence captures most of the behavior; the initial density dependence can be transformed into an initial dependence on s^+ as is described in Ref. [18].

In the case of propane [18], such an initial density term in the entropy scaling framework was included. But it was not included for Lennard-Jones [25] or alkanes [21]. The correlation proposed in this work is able to represent the most accurate low-density data to within its experimental uncertainty without an explicit initial density term.

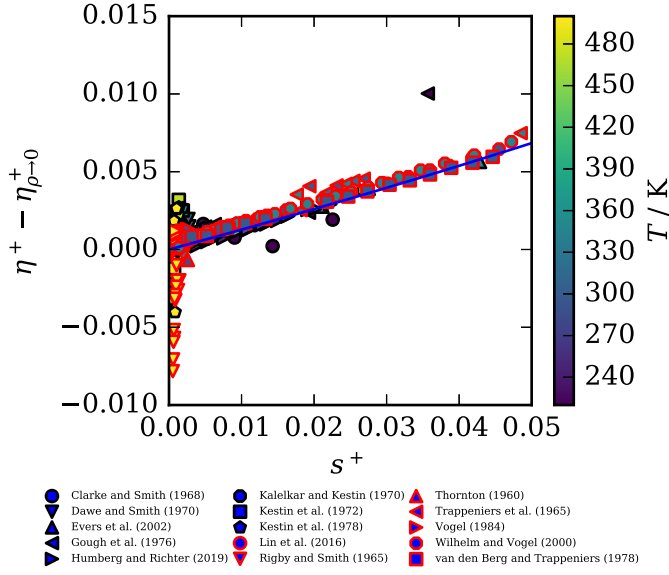


Fig. 7 Scaled residual viscosity in the low-density gas. The solid curve is that of the empirical correlation for Lennard-Jones [25]. Markers in the legend match the shape and outline of the colored markers. Sources as listed in Table 1; some primary datasets are liquid-phase only.

3.3 Formulation

The correlation is therefore given by

$$\eta = \frac{\rho_N^{2/3} \sqrt{mk_B T}}{(s^+)^{2/3}} (1.05 \eta_{\text{LJ, res}}^+ + \eta_{\rho \rightarrow 0}^+(T)) \quad (12)$$

with

$$\eta_{\text{LJ, res}}^+ = -1 + \sum_{i=1}^4 d_i (s^+)^i \quad (13)$$

and with the coefficients for the scaled residual contribution from Lennard-Jones (from Ref. [25]), $d_1 = 0.125364$, $d_2 = 0.220795$, $d_3 = -0.0313726$, and $d_4 = 0.00313907$. The contribution $\eta_{\rho \rightarrow 0}^+$ comes from Eq. (9). A complete implementation of the model is given in Fig. 8 in the Python programming language. To ensure reproducibility, the viscosity values were calculated with the script used to generate Fig. 8 and calculated values are presented in Table 3.

```

import math

import CoolProp.CoolProp as CP

fld = 'Krypton' # or 'REFPROP::KRYPTON' if you have REFPROP installed
k_B = 1.380649e-23 # J/K
R = CP.PropsSI('gas_constant', fld)
N_A = R/k_B
M = 0.083798 # kg/mol
m = M/N_A # kg per atom

def get_eta0(T):
    """ From https://doi.org/10.1063/1.5125100 """
    a = [0, 9.1297123e-1, -1.0014703e-1, -2.4547423e-2, 3.1450093e-2, -4.4562573e-3,
          -4.5112433e-3, 2.2375443e-3, -1.4554223e-4, -2.0063853e-4, 8.3412883e-5,
          -1.5202363e-5, 1.1590853e-6]
    eta_ref = 25.3062/1e6 # Pa*s
    T0 = 298.15 # K
    N = len(a)
    r = eta_ref*math.exp(sum([a[i]*math.log(T/T0)**i for i in range(N)]))
    return r

def get_etaplus_dil(T):
    rhomolar = 1e-10 # something very close to zero
    B2 = CP.PropsSI('Bvirial', 'T', T, 'Dmolar', rhomolar, fld) # m^3/mol
    dB2dT = CP.PropsSI('dBvirial_dT', 'T', T, 'Dmolar', rhomolar, fld) # m^3/mol
    Theta2 = (B2 + T*dB2dT)/N_A # m^3/molecule
    return get_eta0(T)/(m*k_B*T)**0.5*(Theta2)**(2/3)

def get_etaplus_res(splus):
    cLJ = [0, 0.125364, 0.220795, -0.0313726, 0.00313907]
    f = 1.05 # Scale factor on Lennard-Jones correlation
    LJ_contrib = math.exp(sum([cLJ[i]*splus**i for i in range(5)]))-1.0
    return LJ_contrib*f

def get_eta(*, T, rhomolar):
    """
    Return viscosity of Krypton, in Pa*s
    T: Temperature, in Kelvin
    rhomolar: Density, in mol/m^3
    """
    rhoN = rhomolar*N_A
    splus = -CP.PropsSI('Smolar_residual', 'T', T, 'Dmolar', rhomolar, fld)/R
    etaplus_res = get_etaplus_res(splus)
    etaplus = etaplus_res + get_etaplus_dil(T)
    return rhoN**(2/3)*(m*k_B*T)**0.5*etaplus/splus**(2/3)

if __name__ == '__main__':
    T = 400 # K
    rhomolar = 6000.0 # mol/m^3
    print(get_eta(T=T, rhomolar=rhomolar))

```

Fig. 8 Implementation of the model in the Python programming language

Table 3 Check values to ensure the proper implementation of the model. Values were calculated with the implementation in Fig. 8. The large number of significant digits are to ensure proper implementation and are not indicative of uncertainty in the quantity.

T / K	$\rho / \text{mol/m}^3$	$\eta / \mu\text{Pa s}$
200	10^{-6}	17.33865170451214
200	13020.0	56.4476422453026
298.15	10^{-6}	25.306200000810886
400	10^{-6}	32.795558620965195
400	13020.0	64.8014771396677

The deviations between the model and the experimental data are shown in Fig. 9 as a function of s^+ . Reading the deviation plot from left-to-right we have the

dilute-gas limit, gaseous, intermediate, and dense liquid states. The value of s^+ at the critical point is representative of crossing from gas-like to liquid-like dynamics [75].

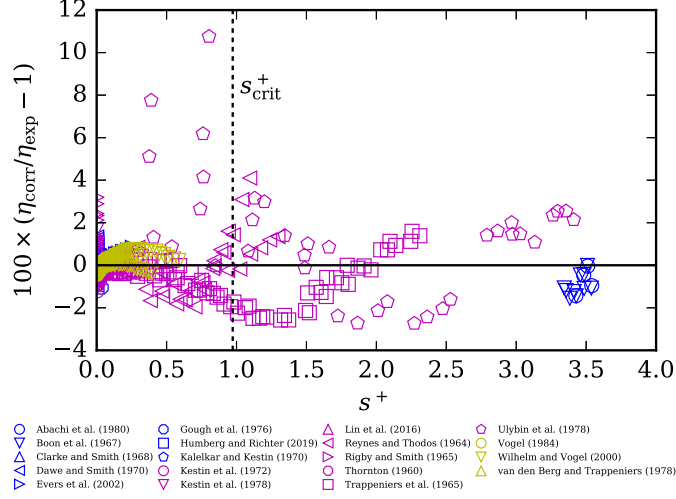


Fig. 9 Deviations in η as a function of s^+ . Sources as listed in Table 1.

Table 4 Deviation statistics per source from the model in this work.

author(year)	AAD	BIAS	MAX
Humberg and Richter (2019)	0.05	0.02	0.16
Lin et al. (2016)	0.04	-0.02	0.09
Evers et al. (2002)	0.36	0.32	0.84
Wilhelm and Vogel (2000)	0.20	0.08	0.74
Vogel (1984)	0.29	-0.29	0.43
Abachi et al. (1980)	0.73	-0.73	1.44
Kestin et al. (1978)	0.87	-0.49	1.67
Ulybin et al. (1978)	2.40	1.43	10.76
van den Berg and Trappeniers (1978)	0.27	0.22	0.74
Gough et al. (1976)	0.29	-0.28	1.05
Kestin et al. (1972)	0.63	-0.51	1.21
Dawe and Smith (1970)	0.72	0.34	1.33
Kalelkar and Kestin (1970)	0.42	-0.35	0.93
Clarke and Smith (1968)	0.26	-0.09	0.52
Boon et al. (1967)	0.96	-0.96	1.55
Rigby and Smith (1965)	1.44	1.42	3.20
Trappeniers et al. (1965)	0.80	-0.61	2.57
Reynes and Thodos (1964)	1.00	-0.05	4.10
Thornton (1960)	0.42	0.42	0.42

Table 4 shows the statistics of fit for each of the primary datasets. The deviation terms are defined by

$$\text{ERR}_k = 100 \times (\eta_{\text{corr},k} / \eta_{\text{exp},k} - 1) \quad (14)$$

$$\text{AAD} = \text{mean}(\text{abs}(\text{ERR})) \quad (15)$$

$$\text{BIAS} = \text{mean}(\text{ERR}) \quad (16)$$

$$\text{MAX} = \text{max}(\text{abs}(\text{ERR})) \quad (17)$$

with the subscript k corresponding to the index of the k -th data point in the ERR array.

Fig. 10 shows the low-density data, indicated by data points with $s^+ < 0.01$. Aside from a deviating dataset from Rigby and Smith [63], the deviations show only a weak temperature dependence. The most accurate datasets carried out in approximately the last 20 years (Humbert and Richter [49], Lin et al. [50], Evers et al. [51], Wilhelm and Vogel [52]), are represented within 0.84% at worst, although the measurements only cover the temperature range 243 K to 473 K (representing much of the technically relevant temperature range).

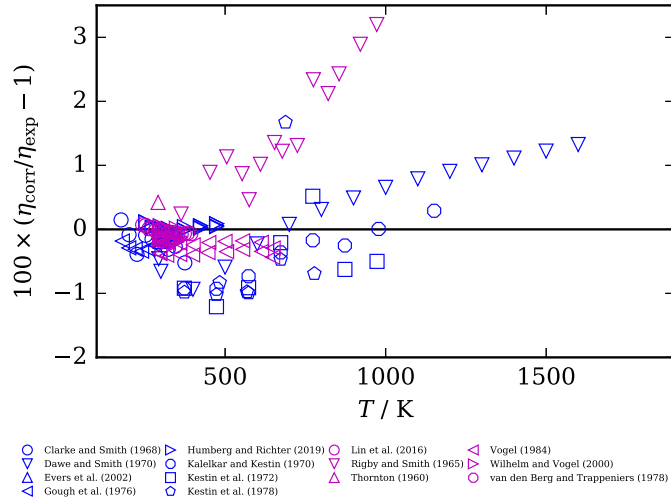


Fig. 10 Deviations in η in the gas phase for data points with $s^+ < 0.01$. Sources as listed in Table 1.

The current reference model for the viscosity of krypton is the ECS model described in Ref. 10. The deviations calculated by REFPROP 10 are shown in Table 5. The ECS-based model provides a better representation of the data of Ulybin et al. [54] than the current model, but nearly all other datasets are better represented by the present model. The central 95 percentiles of the signed relative error are $[-4.6, 2.0]$ with the ECS model and $[-2.2, 2.5]$ with the model presented in this work.

Table 5 Deviation statistics per source with the extended corresponding states model in REFPROP 10 [19].

author(year)	AAD	BIAS	MAX
Humberg and Richter (2019)	0.52	-0.42	1.48
Lin et al. (2016)	0.29	-0.19	0.74
Evers et al. (2002)	0.24	-0.20	0.51
Wilhelm and Vogel (2000)	0.69	-0.67	3.73
Vogel (1984)	1.20	-1.20	2.39
Abachi et al. (1980)	2.13	2.13	2.92
Kestin et al. (1978)	1.92	-1.92	3.05
Ulybin et al. (1978)	1.78	-0.12	4.38
van den Berg and Trappeniers (1978)	0.11	0.04	0.23
Gough et al. (1976)	0.54	-0.54	2.07
Kestin et al. (1972)	2.17	-2.17	3.39
Dawe and Smith (1970)	2.14	-2.14	3.01
Kalelkar and Kestin (1970)	2.21	-2.21	3.07
Clarke and Smith (1968)	0.41	-0.40	1.14
Boon et al. (1967)	1.69	1.69	2.95
Rigby and Smith (1965)	0.47	-0.44	1.24
Trappeniers et al. (1965)	2.54	-2.54	4.74
Reynes and Thodos (1964)	3.17	-3.09	6.62
Thornton (1960)	0.47	0.47	0.47

The box of isomorph breakdown is bounded by temperatures from the critical temperature to 298 K and with s^+ in (0.5, 1.0). Outside this box, the expanded uncertainty (at the 95% confidence level) is approximately 2%, with expanded uncertainties decreasing to less than 0.1% in the dilute gas limit. Inside the box of isomorph breakdown, the expanded uncertainty is greater, conservatively up to 5 % (as would be consistent with the scatter seen in this region for propane [18]), but only a few experimental data points are present in this region from a group with an otherwise reliable track record. The correlation can be expected to systematically over-predict the viscosity in the isomorph breakdown region as it tracks the high-temperature nearly monovariate curve.

For Lennard-Jones, the existing molecular dynamics data extend to physically unreasonably high temperatures, and so we can have good confidence that so long as the values of s^+ (and density) obtained from the EOS are reasonable, so should the values for viscosity obtained from the correlation. This highlights the power of the entropy scaling approach: a very simple function of a single independent variable can be used to represent the residual (and macroscopically scaled) viscosity surface.

4 Conclusions

In this work a reference correlation for the viscosity of krypton has been presented with one adjustable parameter in the liquid phase as a consequence of scaling the residual contribution of the Lennard-Jones fluid. The correlation is non-iterative and can be implemented in any programming language. The correlation is valid from 70 K to 5000 K in the dilute gas, and from 115.775 K to 750 K in the fluid phase, with a pressure limit equal to that of the melting curve.

One point which was only lightly touched upon in this work thus far is the quality of the equation of state, and especially how accurately it represents the residual entropy. For the dilute gas, it was already shown (Fig. 4) that Θ_2 is not correct at low temperatures, and this is the leading density dependence of s^+ at low density. In a virial expansion, $s^+ = \Theta_2\rho_N + \frac{1}{2}\Theta_3\rho_N^2 + \dots$ (see the supporting information of Ref. [18]). For dense states, further investigations of the EOS is required. In the case of propane, it was shown that changing the EOS to a slightly less accurate (and older) multiparameter EOS has only a very modest impact on the accuracy of the correlation [18]. In the case of krypton, we expect that the critical region could be a region where changes in the EOS are particularly strongly felt.

There exist some significant holes in the experimental data coverage for krypton, especially in the temperature range from the critical temperature of approximately 200 K to room temperature over the full density range. In this region only one dataset is available in the literature. New high accuracy measurements in this region would allow for a more reliable representation of the viscosity. These measurements would also allow for investigation of the ways in which krypton is (and is not) well represented by the entropy scaling framework. And in particular, what sort of approach (empirical, or ideally physics-based) is needed to repair the breakdown of the scaling.

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Data Accessibility Statement

In order to ensure reproducibility of the results, the supplementary material includes primary data in tabular form.

Appendix A: Derivation of high-temperature limit

The second virial coefficient is defined by

$$B_2 = -2\pi \int_0^\infty [(\exp(-\beta V(r)) - 1)r^2]dr \quad (18)$$

where $V(r)$ is the potential, and $\beta = 1/(k_B T)$. The product βV is dimensionless. Any potential that is finitely valued in the entire domain of integration has the infinite temperature limit

$$\boxed{\lim_{\beta \rightarrow 0} n_{\text{eff}} = \frac{3}{2}} \quad (19)$$

where

$$n_{\text{eff}} = -3 \frac{B_2 - \beta \frac{dB_2}{d\beta}}{\beta^2 \frac{d^2 B_2}{d\beta^2}} \quad (20)$$

The derivation begins with substitution for B_2 , yielding

$$n_{\text{eff}} = -3 \frac{-2\pi \int [(\exp(-\beta V) - 1)r^2]dr - 2\pi \int \beta V \exp(-\beta V)r^2 dr}{-2\pi \int (\beta V)^2 \exp(-\beta V)r^2 dr} \quad (21)$$

and after joining terms

$$n_{\text{eff}} = -3 \frac{\int [(1 + \beta V) \exp(-\beta V) - 1]r^2 dr}{\int (\beta V)^2 \exp(-\beta V)r^2 dr} \quad (22)$$

which has an indefinite form for $\beta \rightarrow 0$. In this case, two applications of de l'Hôpital's rule are required, such that

$$\lim_{\beta \rightarrow 0} n_{\text{eff}} = -3 \lim_{\beta \rightarrow 0} \frac{\frac{d^2}{d^2\beta^2} \int [(1 + \beta V) \exp(-\beta V) - 1]r^2 dr}{\frac{d^2}{d^2\beta^2} \int (\beta V)^2 \exp(-\beta V)r^2 dr} \quad (23)$$

or

$$\lim_{\beta \rightarrow 0} n_{\text{eff}} = -3 \lim_{\beta \rightarrow 0} \frac{\int [V^2(\beta V - 1) \exp(-\beta V)]r^2 dr}{\int V^2(V^2\beta^2 - 4V\beta + 2) \exp(-V\beta)r^2 dr}, \quad (24)$$

and finally yielding

$$\lim_{\beta \rightarrow 0} n_{\text{eff}} = -3 \lim_{\beta \rightarrow 0} \frac{\int -V^2 r^2 dr}{\int 2V^2 r^2 dr} = \frac{3}{2} \quad (25)$$

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