GENERALIZED THERMODYNAMIC BEHAVIOR FOR THE GASEOUS AND LIQUID STATES I. THERMAL CONDUCTIVITY

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Abstract—Thermal conductivity measurements available in the literature, for 23 fluids in their dense gaseous and liquid states, have been used to develop the generalized relationship for excess thermal conductivity. Calculated thermal conductivities relating to these 23 substances which include monatomic, diatomic and polyatomic fluids and hydrocarbons of all types have been compared with corresponding experimental measurements to produce an overall average deviation of 3.87% (1111 points) for all fluid conditions through the compressed liquid state.

INTRODUCTION

The treatment relating to the thermal conductivity of fluids in their dilute gaseous state has been difficult to accommodate in a generalized manner. This difficulty also extends to include the behavior of the dense gaseous and liquid regions. In this state of aggregation, the prediction of this transport property is complicated by multiple collisions and forces of attraction that are complex in nature. In particular, this argument extends to liquid systems consisting of polyatomic molecules since no satisfactory statistical solution is yet available to account for their behavior in this dense state of aggregation. In this context, the prediction of thermal conductivity is beset with difficulties because the individual translational, rotational and vibrational modes of energy transfer are not well understood under varying temperature and pressure conditions.

Attempts to treat thermal conductivity in a generalized manner are reported by Gamson [1] who developed, from measurements available in the fiterature, a corresponding states correlation in which the reduced thermal conductivity, K/K_c , is plotted against reduced temperature for parameters of reduced pressure. Comings and Nathan [2] combined the Enskog relationships for viscosity and thermal conductivity to obtain an expression for the ratio, K/K° , in terms of the viscosity ratio, μ/μ° and y, the Enskog modulus. In their final relationship, the effects of the internal degrees of freedom of the molecules was neglected. Stiel and Thodos [3] utilized a dimensional analysis approach which involves the use of the critical constants and showed that excess thermal conductivity depends on reduced density and the critical compressibility factor. More specifically, they were able to produce a unique expression for dense gases and liquids relating ($K-K^{\circ}$) λZ_c^5 versus ρ_R for 20 substances which included monatomic, diatomic and polyatomic molecules and hydrocarbons of all types. In this expression, $\lambda = T_c^{16}M^{1/2}/P_c^{2/3}$. Their correlation extends up to reduced densities of $\rho_R = 2.80$ and is restricted in application to nonpolar substances. However, the generalized behavior of anomalous thermal conductivity enhancement appearing near the critical point has been separately investigated in the previous works [4,5].

DILUTE GASEOUS STATE

For the thermal conductivity of the dilute gaseous state of rigid spherical molecules having negligible attractive forces of interaction, molecular theory has been applied to the development of the relationship [6],

$$K^* = 19.981 \times 10^{-5} \frac{\sqrt{T/M}}{\sigma^2 Q^{(2,2)^*}(T^*)}.$$
 (1)

This relationship predicts thermal conductivities accuately for monatomic gases and is consistent with the simple kinetic theory which considers only the transfer of translational energy of such rigid spherical molecules. To extend the application of thermal conductivity to polyatomic gases in their dilute state, Roy and Thodos [7] applied a dimensional analysis approach and showed that (4)

$$K^* \lambda = \beta Z_c^m T_R^n \tag{2}$$

where the thermal conductivity parameter, $\lambda = T_c^{1/6} M^{1/2} / P_c^{2/3}$. From the analysis of data available in the literature, they concluded that m = 0 and that $K^* \lambda$ is only a function of T_R . However, this dependence was found to relate uniquely to molecular complexity.

For the monatomic gases, which possess only translational degrees of freedom, Equation (2) was found to be,

$$(K^*\lambda)_m = 9.96 \times 10^{-5} [\exp(0.0464 T_R) - \exp(-0.2412 T_R)]$$
(3)

for temperature up to $T_R \approx 20$. Equation (3) reproduced experimental measurements for neon, argon, krypton and xenon with an average deviation of 2.8% (219 points). However, in order to take into account the transfer of energy between translational and internal degrees of freedom in polyatomic gases, Roy and Thodos [7] extended their treatment beyond monatomic gases and presented for diatomic and triatomic gases the relationships,

 $(K^*\lambda)_d = 1.10 (K^*\lambda)_m^{1.10}$ (diatomic gases)

and

$$(K^*\lambda)_t = 1.02 (K^*\lambda)_m^{1.46}$$
 (triatomic gases). (5)

Equation (4) has been used to calculate thermal conductivities for the diatomic gases, nitrogen, carbon monoxide, nitric oxide, oxygen, fluorine, hydrogen chloride and hydrogen bromide which, when compared with corresponding experimental measurements, produced an average overall deviation of 4.3% (282 points). For the triatomic gases, carbon dioxide, sulfur dioxide, nitrous oxide and water vapor, Equation (5) reproduced experimental measurements with an overall average deviation of 4.64% (242 points).

For the complex nature of hydrocarbons that include normal aliphatics and their isomers, olefins, acetylenes, naphthenes, and aromatics, Roy and Thodos [8] utilized a group contribution approach to account for the sum of the rotational and vibrational contributions of these substances and represented them as the difference between the total and translational contributions.

$$(K^*\lambda) - (K^*\lambda)_t = (K^*\lambda)_\tau - (K^*\lambda)_v.$$
(6)

This approach was utilized by them to develop a method for predicting thermal conductivities for 27 hydrocarbons of all degrees of complexity with an overall average deviation of 2.1% (109 points). Using the same arguments, their approach was extended [9] to the development of a method for the treatment of organic compounds including alcohols, esters, ketones, aldehydes, halides, amines and cyclics. For 52 organic compounds, a comparison between calculated and experimental values produced an average deviation of 3.0% (291 points).

BASIC CONSIDERATION FOR THE NON-DILUTE GASEOUS STATE

An attempt to account for the excess thermal conductivity, K-K*, of nonpolar fluids of all degrees of molecular complexity was reported by Stiel and Thodos [3]. Their approach involved the use of a dimensional analysis, based on the critical constants of a fluid, to show that

$$(K-K^*)\lambda Z_c^s = f(\rho_R).$$
⁽⁷⁾

This dependence was found to apply to monatomic, diatomic, and triatomic fluids and hydrocarbon of all types in their dense gaseous and saturated liquid states for temperatures above the normal boiling point. In their study, because of the lack of experimental liquid measurements, not much attention was directed to the behavior of thermal conductivity for temperatures below their normal boiling point and particularly those approaching the triple point region and also for conditions associated with the compressed liquid state.

A projection to transcend the inability to predict in generalized manner thermal conductivities in the highly compressed region, and particularly in the proximity of the liquid-solid transition state, requires a close scrutiny of the behavior of this transport property with particular emphasis placed at this extreme state of liquid aggregation. Experimental measurements are now becoming available and report thermal conductivities that extend as low as the triple point and which also accommodate the compressed liquid region. A dimensional analysis, based on the triple point values of a fluid, presents an alternate approach that may have more significant implications to the generalized treatment of thermal conductivity than has been possible with that using critical constants. Thus, the excess thermal conductivity may be related to temperature and volume as follows:

$$K - K^* = \alpha T^a T^b_t M^c \nu^a_{tt} R^e \nu^{J}.$$
⁽⁸⁾

Using mass-length-time-temperature dimensions. # follows from Equation (8) that,

$$(K - K^* \cdot \Psi = \beta \frac{\tau^a}{\omega'} \tag{9}$$

where $\Psi = M^{1-2} v_{tt}^{2+3} / T_t^{1+2}$, $\beta = \alpha R^{3-2}$, $\tau = T / T_t$ and $\omega = \rho / \rho_{tt}$. For this state of liquid aggregation, it is interesting to draw a parallel line of reasoning between that proposed by Andrade [10] for viscosity and the present study dealing with thermal conductivity. According to Andrade, the molecules in liquids may be regarded as executing vibrations just as they do in the solid state. The movement of the equilibrium positions of these molecules has been shown to be very slow and need not be taken into account if interest is not directed to such phenomena as self-diffusion and viscosity. Following the procedure outlined by Andrade for viscosity, it can be inferred that the thermal conductivity of liquids becomes

$$K = \frac{4 \varkappa \nu}{\sigma} \tag{10}$$

where \mathbf{x} is the Boltzmann constant, \mathbf{v} is the frequency of vibration at the melting point and $\sigma = (v_{lt}/N)^{1/3}$, the average distance between molecular centers. For the frequency of vibration, Andrade presented the expression

$$\nu = 2.8 \times 10^{-12} \sqrt{\frac{T_t}{M v_{tt}^{2.3}}} \,. \tag{11}$$

Combining Equations (10) and (11) with $\sigma = (v_{\#}/N)^{1/3}$, it can be shown that

$$K\frac{M^{1/2}v_{ll}^{2/3}}{T_l^{1/2}} = 3.1 + 10^{-3}.$$
 (12)

The approximation given by Equation (12) applies at the triple point and is closely related to the application involving spherical molecules. However, this expression cannot be applied for conditions removed from the triple point region and also for nonspherical complex molecules.

DENSE GASEOUS AND LIQUID STATES

Experimental information relating to the behavior of thermal conductivity in the dense gaseous and liquid states is now becoming available for a limited number of fluids. In this connection, experimental thermal conductivities for the dense gaseous and compressed liquid state of oxygen are reported by Roder [11] who also investigated the behavior of propane in its highly compressed liquid state [12]. Along these lines, Menashe and Wakeham [13] reported thermal conductivity measurements for the liquid state of *n*-heptane and also for *n*-nonane and *n*-undecane [14], each for pressures up to 500 MPa (4935 atm). The results for *n*-heptane are expressed as excess thermal conductivity, $(K-K^*)\Psi$ versus ω , the density normalized with the liquid density at the trip e-point. The

experimental measurements for oxygen, propane, *n*-nonane and *n*-decane exhibited patterns of behavior similar to that found for *n*-heptane. The behavior of all these fluids indicates that thermal conductivity continues to be strongly dependent on density, and weakly dependent on temperature.

The dependence of $(K-K^*) \Psi$ on the both temperature and density is consistent with the results of the dimensional analysis expressed by Equation (9). However, the term, τ^{a}/ω' , is not so straightforward, but turns out to be more complex in form as depicted from actual measurements. A careful coordination of both temperature and density introduces *x*, the single variable combining both of these properties as follows:

$$x = \omega \tau^{a \omega^n}$$
(13)

where *a* and *n* are constants to be established from experimental measurements. Using *a* = 0.08527 and *n* = 1.8158 determined from the nonlinear regression analysis, the three isotherms combine to the unique dependence of $(K-K^*) \Psi$ versus *x*, the single densitytemperature variable. The application of this variable using *a* = 0.08527 and *n* = 1.8158 extends as well to the application of the data available for oxygen and the normal hydrocarbons, *n*-propane, *n*-nonane and *n*undecane to produce the unique, but different relationships of $(K-K^*) \Psi$ versus *x* for each of these fluids.

Additional substances, varying in molecular complexity from monatomic, diatomic and polyatomic configurations such as cyclohexane, benzene and toluene, were included in this study. Experimental thermal conductivities for the dense gaseous and liquid state of these substances obtained from literature sources were presented in Table 2, which altogether included 23 fluids of an essentially nonpolar nature. Excess thermal conductivities expressed as $(K-K^{-})\Psi$ when related to *x*, the combined density-temperature variable, produced for each fluid continuous and unique relationships for the dense gaseous, saturated liquid and compressed liquid regions. The general form of these relationships can be expressed by the following functional dependence.

$$(K - K^*) \Psi = \exp[\alpha x^m + \beta x^n] - 1 \tag{14}$$

where α , β , *m* and *n* are specific constants associated with this functional dependence for each substance. Table 1 includes the values of these constants for each fluid obtained from a nonlinear regression analysis. This table also includes other basic constants related to these fluids.

GENERALIZED TREATMENT FOR THE DENSE GASEOUS AND LIQUID STATES

Table 1. Basic constants associated with the fluids of this study

		0	ر م	ر د	11	VII	۲	Ð	ø	β	E	r L
Monatomic												
Neon	20.183	44.45	26.86	41.79	24.54	16.21	0.9428	5.8087	4.89682	0.14273	0.12996	8.13162
Argun	39.948	150.86	48.34	74.56	83.79	28.21	1.0989	6.3986	4.96497	0.20647	0.37683	5.54671
Krypton	83.80	209.41	54.18	92.29	115.94	34.31	1.5580	8.9768	4.74192	0.42203	0.33825	2.61236
Xenon	131.30	289.76	57.64	119.50	161.36	42.66	1.9754	11.0132	4.89735	0.46204	0.33161	4.87932
Diatomic												
Nitrugen	28.013	126.2	33.54	89.20	63.14	32.29	1.1398	6.7542	5.42691	-0.08389	0.44561	3.05828
Oxygen	32.00	154.58	49.77	73.49	54.363	24.81	0.9666	6.5264	5.21947	0.42877	0.33874	2.96872
Hydrocarbons												
Methane	16.043	190.555	45.39	100.00	90.66	35.37	0.7551	4.5327	5.02870	0.34608	0.40150	3.97308
Ethylene	28.054	282.40	50.50	123.60	104.00	42.62	0.9930	6.3371	5.22406	1.12093	0.19682	4.99349
Ethane	30.070	305.4	48.20	148.00	89.88	45.58	1.0744	7.3805	4.53761	1.69699	0.21297	1.80233
Propane	44.097	369.8	41.90	203.0	85.44	60.28	1.4748	11.0448	5.14084	1.16202	0.32517	1.57441
<i>n</i> -Butane	58.124	425.18	37.47	254.91	134.86	78.44	1.8670	12.0299	4.84079	1.43499	0.46469	0.96412
n-Pentane	72.151	469.6	33.30	304.0	143.40	95.10	2.2879	14.7786	5.59704	0.79278	0.55707	0.55706
<i>n</i> -Hexane	86.178	507.4	29.30	370.0	177.84	113.45	2.7585	16.3137	5.62357	0.80925	0.26746	2.78701
Cyclohexane	84.16	553.2	40.00	308.05	279.83	106.30	2.2473	12.3063	0.48930	5.39987	-1.29472	0.73229
Benzene	78.115	562.09	48.34	258.66	278.693	87.31	1.9134	10.4195	0.19131	3.97859	-0.94246	1.44503
<i>n</i> -Heptane	100.198	540.3	27.00	431.94	182.57	129.20	3.1741	18.9331	6.97530	-0.48510	0.53518	1.74200
Toluene	92.141	591.79	40.50	315.55	178.169	94.55	2.3584	14.9251	7.28375	-0.93273	0.72158	3.48830
n-Octane	114.232	568.8	24.50	492.0	216.38	150.38	3.6471	20.5469	7.07384	-0.51786	0.67427	2.97773
<i>n</i> -Nonane	128.250	593.8	22.53	552.63	219.66	165.58	4.1159	23.0405	6.07777	0.55438	0.43891	0.43904
<i>n</i> -Decane	142.286	617.6	20.80	603.0	243.51	184.25	4.6025	24.7502	5.92427	0.82000	0.48045	0.53398
<i>n</i> -Undecane	156.313	638.8	19.40	660.0	247.6	203.28	5.0814	27.4696	6.07407	0.69120	0.43407	0.43407
Miscellaneous												
Carbon Dioxide	44.01	304.19	72.85	94.04	216.55	37.37	0.9863	5.0391	4.74710	0.54169	0.27829	2.70549
Carbon Tetra-	153.823	556.40	45.00	276.00	250.00	91.96	2.8115	115.9811	5.30193	0.74353	0.07943	4.03077

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Compd.	ζ	T _{Rt}	Z _c (lit.)	Z _c (calc'd)	Pts	% Dev.	Lit.
Monatomic							. ———
Neon	1.5615	0.5521	0.298	0.298	6	1.58	[16]
Argon	1.4984	0.5554	0.293	0:293	169	4.47	[17]
Krypton	1.4985	0.5537	0.292	0.293	147	4.46	[18]
Xenon	1.4313	0.5569	0.290	0.289	77	4.39	[18]
Diatomic							(10)
Nitrogen	1.3500	0.5003	0.290	0.289	17	2.13	[19]
Oxygen	1.2365	0.3517	0.288	0.289	116	1.57	[11]
Hydrocarbons							11
Methane	1.4069	0.4758	0.289	0.294	151	8.30	[20]
Ethylene	1.1283	0.3683	0.276	0.278	7	0.69	[21]
Ethane	1.0678	0.2943	0.274	0.276	65	6.06	[22]
Propane	0.9940	0.2310	0.271	0.270	70	1.42	[12]
n-Butane	1.0140	0.3172	0.274	0.269	6	0.91	[23]
<i>n</i> -Pentane	0.9695	0.3054	0.269	0.265	16	1.29	[23]
n-Hexane	0.9567	0.3505	0.263	0.261	13	0.64	[23]
Cyclohexane	1.1380	0.5058	0.277	0.271	49	3.97	[24]
Benzene	1.1502	0.4958	0.271	0.273	12	1.75	[25]
<i>n</i> -Heptane	0.9480	0.3379	0.260	0.260	50	3.45	[13]
Toluene	0.9718	0.3011	0.263	0.265	16	0.86	[23]
n-Octane	0.9183	0.3804	0.255	0.256	11	0.31	[23]
<i>n</i> -Nonane	0.9141	0.3699	0.250	0.256	64	3.66	[14]
<i>n</i> -Decane	0.8696	0.3943	0.245	0.250	9	0.46	[23]
<i>n</i> -Undecane	0.8813	0.3876	0.240	0.252	51	3.19	[14]
Miscellaneous							1
Carbon Dioxide	1.4202	0.7119	0.274	0.274	79	1.22	[26]
Carbon Tetrachloride	1.0685	0.4493	0.272	0.268	9	2.28	[25]

Table 2. Basic parameters and deviations resulting from eq. (17)

The nonlinear relationships of $(K-K^*)$ ψ versus x, begin from the origin and increase exponentially to approach for each fluid the respective freezing curve, where the thermal conductivity remains finite. The monatomic fluids, neon, argon, krypton and xenon exhibit the least dependence of $(K-K^*) \Psi$ on x, whereas for the structurally complex molecules, this dependence increases more rapidly, as exhibited, by the heavier hydrocarbons. In general, with increasing molecular complexity, the critical compressibility factor, Z_c is found to decrease progressive y. However, besides Z_{cr} a strong inference exists that an additional parameter also plays a role in defining the $(K-K^{\circ})\Psi$ versus x behavior of each fluid. This additional parameter has been taken to represent the volume increase as the solid melts at the triple point and has been designated as $\varepsilon = v_{ll}/v_{sl}$. Thus, the combined influence of Z_{c} and ε bears directly on the characterizing nature of the fluid and has been defined as.

$$\zeta = f(Z_c, \varepsilon) \tag{15}$$

where ζ can be applied to define the global variable as follows:

$$g = \frac{x}{\zeta^{x^{\tau}}}.$$
(16)

This variable is an all inclusive factor that represents in a combined manner the influence of density and temperature along with the parameters ε and Z_c needed to characterize the physical nature of the fluid. Based on these premises, the variable *g* assumes a universal significance capable of defining excess thermal conductivity in a manner similar to Equation (14) as follows:

$$(K - K^*) \Psi = \exp[ag^n + bg^\nu] - 1 \tag{17}$$

where a, b, η , and ν are assumed to be constants applicable to all fluids. To establish these constants, the fluid characterizing parameter, ζ , and exponent, r, must first be determined individually for each substance. This approach requires the simultaneous solutions for six unknowns. A nonlinear regression analysis of the experimental values $(K-K^*)\Psi$, density and temperature, for the 23 fluids included in this study produced for each parameter of Equation (17), the following values:

<i>a</i> == 3.5761	$\eta = 0.1602$
b=2.6963	v = 0.9941

and the value r = 0.13963 needed to define *g* in Equation (16). On the other hand, the fluid characterizing parameter ζ was found to be constant and unique to a fluid, but to vary from substance to substance. The best values of ζ obtained from this treatment are included in Table 2 and, in general, decrease with molecular complexity. The highest value was found for neon, $\zeta = 1.56147$ and the lowest for *n*-decane, $\zeta = 0.86958$.

ESTIMATION OF PARAMETER ζ

The fluid characterizing parameter ζ has been assumed to depend on the critical compressibility factor, Z_c and ε , the volume expansion factor at the triple point, through the relationship,

$$\zeta = AZ_c^c + B\varepsilon^d + CZ_c^f \varepsilon^g \tag{18}$$

where *A*, *B*, *C*, *c*, *d*, *f* and *g* are constants. Since reliable values for volume expansion factors are difficult to obtain, a method has been devised for the estimation of this factor using reasonable thermodynamic arguments. To predict ε , the molar volumes of the solid and liquid states at the triple point must be available. The liquid molar volume can be calculated by applying the Rackett equation [15] at the triple point to yield,

$$v_{tt} = v_c Z_c^{1-T_R} t^{0.286} \tag{19}$$

The solid molar volume at the triple point relates linearly with b, the modified van der Waals covolume parameter as follows:

$$v_{si} = kb \tag{20}$$

where the proportionality constant has been found to be k > 1.20 and the covolume parameter is defined through the modified van der Waals equation of state.

$$(P \cdot \frac{a}{v^n})(v-b) = RT \tag{21}$$

in which $b = [(n-1)/(n+1)]v_c$ and $n = 2Z_c - \sqrt{4Z_c^2 + 1}$. Therefore it follows that, for the calculation of parameter *b*, all critical constants must be available. With these interlocking relationships, ε , the volume expansion factor at the triple point now can be defined, with the elimination of v_c , as follows:

$$\varepsilon = \frac{Z_c^{(1-T_R)^{-0.286}}}{k(n-1)/(n+1)} .$$
(22)

Through this approach, ϵ is shown to depend upon the two parameters Z_c and T_{R_f} . Values of these parameters are presented in Table 2 for each fluid included in this treatment. A nonlinear regression analysis has been applied, using values of Z_c and T_{R_f} available in the literature, to establish the constants A, B, C, c, d, f and g of Equation (18). The compromising constants of this equations for the 23 fluids were found to be

Since the accuracy of the triple point parameter, T_{R_c} is better known than the corresponding Z_c value, the fluid characterizing parameter ζ , expressed throught the relationship of Equation (18), becomes strongly influenced on the accuracy associated with Z_i . The parameter ξ associated with a fluid represents a compromise obtained from a number of thermal conductivity measurements ranging from low to high densities, which extend to the triple point region, and therefore this parameter for a substance is known better than its corresponding Z_c value. For the sake of comparison, values of Z_c were calculated with Equation (18) and its compromised constants. These calculated Z_c values are included in Table 2, and, for the most part, are in good agreement with corresponding values obtained from the literature. The greater departure noted for the heavier normal hydrocarbons, nnonane, n-decane and n-undecane, is undoubtedly due to the uncertainties of both thermal conductivities and critical constants for these fluids. Values for the global variable, g. corresponding to experimental excess thermal conductivities were calculated using Equation (18) to estimate parameter ξ , and density and temperature to obtain x.

CONCLUSIONS

The procedure outlined through the of Equations (13), (16), (17) and (18) permitted the calculation of excess thermal conductivities, K-K^{*}, which were compared with corresponding experimental measurements for all 23 fluids included in the study. The results of this comparison were summarized in Table 2 and produced an overall average deviation 3.87% (1111 points). This deviation includes fluid states that extend from the dense gaseous state into the saturated

and compressed liquid region. The individual average deviations ranged from 0.31% (11 points) for liquid *n*-octane to 8.30% (151 points) for methane in its dense gaseous and liquid states.

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NOMENCLATURE

- g : global variable
- K : thermal conductivity [cal/sec cm K]
- *K*^{*} : thermal conductivity of dilute gaseous state [cal/ sec cm K]
- M : molecular weight
- P : pressure [atm]
- P_c : critical pressure [atm]
- *T* : temperature [K]
- T_c : critical temperature [K]
- T_R : reduced temperature
- $T_{R_{e}}$: reduced temperature at triple point
- T_t : triple point temperature [K]
- v : molar volume [cm³/mol]
- v_c : critical volume [cm³/mol]
- v_{μ} : liquid molar volume at triple point [cm³/mol]
- v_{st} : solid molar volume at triple point [cm³/mol]
- x =: density-temperature variable
- y =: Enskog modulus
- Z_c : critical compressibility factor

Greek Letters

- ε volumetric expansion factor for melting at triple point
- ζ : fluid characterizing parameter
- * : Boltzmann constant
- λ_{-} : thermal conductivity parameter
- μ : viscosity [g/sec cm]
- μ^{\pm} : viscosity of dilute gaseous state [g/sec cm]
- ρ : density [g/cm³]
- $\boldsymbol{\rho}_{k}$: liquid density at triple point [g/cm³]
- ρ_R : reduced density
- σ : average distance between molecular centers
- τ : normalized temperature
- $\boldsymbol{\psi}_{-}$: thermal conductivity parameter
- ω : normalized density

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