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International Journal of Thermal Sciences 41 (2002) 949–954

International Journal of Thermal Sciences

www.elsevier.com/locate/ijts

Calculation of transport properties of simple dense fluids

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Received 6 June 2001; accepted 7 November 2001

Abstract

The transport properties of noble gases and of some other simple fluids including N_2, O_2, F_2, CO, CO_2, CH_4, CF_2Cl_2, C_2H_6, C_3H_8 and SF_6 have been calculated by the modified Enskog theory. The density dependence of the radial distribution function has been evaluated using an analytical equation of state based on the statistical–mechanical perturbation theory. The resultant transport equation of state needs three temperature-dependent parameters, which can be determined by knowing the intermolecular pair potential, plus the zero-density values of the transport coefficients. Using a simple Lennard–Jones (12-6) potential for the calculation of temperature-dependent parameters and the extrapolation of experimental low-density transport coefficient data to the zero-density limit, one can put the transport equation of state in the predictive mode. In this work the thermal conductivity and the viscosity coefficient of the helium-group gases have been calculated over a wide range of temperatures and pressures. A comparison of the calculated and experimental values of the thermal conductivity and the viscosity yields an average absolute deviation of 2.66% for the thermal conductivity and of 3.03% for the viscosity.

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Keywords: Calculation; Equation of state; Modified Enskog theory; Noble gases; Thermal conductivity; Transport properties; Viscosity coefficient

1. Introduction

The transport properties of fluids are important quantities required in engineering design for production, processing, and fluid transformation. These properties for a dense hard-sphere gas were first formulated by Enskog [1], but there exists no satisfactory theory for real dense gases or liquids. Most of the prediction methods for real fluids are limited to the temperature-dependence of either the dilute gases or liquids at atmospheric pressures [2,3]. Meanwhile there are a few attempts, e.g., Latini and Baroncini [4], to describe the transport properties over a wide range of pressures by adding correction terms. Many correlations have been performed, following the Enskog theory, in the form of reduced density and reduced temperature [5–7]. A compilation of some of these correlations can be found in the book of Reid et al. [3].

Although the original Enskog theory was first proposed for rigid spherical molecules, Enskog pointed out that the transport properties of a dense gas differ from those of a dilute gas because of the instantaneous transfer of energy and momentum over the distance separating the center of mass of one molecule to the center of mass of the other molecules on collision [8]. These contributions have been referred to as collisional transfer contributions. Enskog made two modifications in his original theory to apply it for relatively dense fluids [8,9]. In this method that is called the modified Enskog theory, the actual pressure is replaced by the thermal pressure, in order to take into account the attractive forces between molecules, and the covolume is redefined in terms of the second virial coefficient.

Our knowledge of the transport properties, however, can be simplified when a connection is made with equilibrium properties. In fact, this procedure can sometimes avoid the explicit consideration of a formal theory of transport properties. The transport properties can be expressed in terms of pressure and temperature, or much better in terms of density and temperature, using an accurate equation of state for evaluating the internal pressure. The resulting equation, which describes the temperature and density dependence of the transport phenomena, is called the transport equation of state. As in the thermal equations of state, the transport equations of state may be arbitrarily classified as the equations of the virial type for representation of transport properties, and the cubic equations for extrapolation and prediction. The

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PII: S1290-0729(02)01387-X

Nomenclature

b	covolume	$m^3 \cdot mol^{-1}$
B_2	second virial coefficient	$m^3 \cdot mol^{-1}$
C_v	heat capacity at constant volume	$J \cdot K^{-1}$
D	self diffusion coefficient	$m^2 \cdot s^{-1}$
k	Boltzmann constant	$J \cdot K^{-1}$
m	molecular mass	kg
N_A	Avogadro number	mol^{-1}
p	pressure	$N \cdot m^{-2}$
r	intermolecular distance	nm
R	gas constant	$J \cdot K^{-1} \cdot mol^{-1}$
T	absolute temperature	K
u	potential energy	J
u_0	the repulsive branch of u	J

Greek symbols

α	contribution of repulsive forces to B_2	$m^3 \cdot mol^{-1}$
β	analogous to the van der Waals covolume defined by Eq. (11)	$m^3 \cdot mol^{-1}$
χ	radial distribution function at contact	
ε	potential well-depth	J
Γ	an adjustable parameter in Eq. (12)	
η	viscosity	$N \cdot s \cdot m^{-2}$
λ	thermal conductivity	$W \cdot m^{-1} \cdot K^{-1}$
ρ	molar density	$mol \cdot m^{-3}$
σ	molecular diameter	nm

Subscripts

0	zero-density value

Superscripts

$'$	the contribution from translational degrees of freedom
$''$	the contribution from internal degrees of freedom

purpose of this work is to connect the modified Enskog theory with an accurate equation of state to predict the transport properties of simple fluids over a wide range of temperatures and pressures.

2. Theory

The transport properties of a dense fluid can be related to that of the dilute gas by Enskog theory. For a dense gas of rigid spherical molecules, the pertinent expression for the viscosity coefficient is as follow [8,9]:

$$\eta = \frac{\eta_0}{\chi}\left[1 + \frac{4}{5}b\rho\chi + 0.761(b\rho\chi)^2\right] \tag{1}$$

where η_0 is the dilute-gas viscosity coefficient, ρ is the molar density, $b = 2\pi N_A\sigma^3/3$ is the covolume, σ is the molecular diameter, N_A is the Avogadro number, and χ is the value of equilibrium radial distribution function at a distance σ from a center of an individual molecule. The expression for thermal conductivity takes a similar form to Eq. (1), but for polyatomic molecules the contribution from both internal and translational degrees of freedom should be taken into account, i.e.,

$$\lambda = \frac{\lambda_0'}{\chi}\left[1 + \frac{6}{5}b\rho\chi + 0.755(b\rho\chi)^2\right] + \frac{\lambda_0''}{\chi} \tag{2}$$

with

$$\lambda_0' = \frac{5}{2}\eta_0\frac{C_v'}{m} \tag{3}$$

$$\lambda_0'' = \rho D_0 N_A \frac{C_v''}{m} \tag{4}$$

$$\lambda_0 = \lambda_0' + \lambda_0'' \tag{5}$$

and

$$C_v = C_v' + C_v'' \tag{6}$$

where λ_0 and D_0 are the dilute gas thermal conductivity and self diffusion coefficient, respectively, C_v is the heat capacity at constant volume, m is the molecular mass, and primed and double primed quantities represent the contributions from translational and internal degrees of freedom, respectively. The dilute-gas expressions given in the fourth Enskog approximation for η_0 and λ_0 of hard spheres read as:

$$\eta_0 = 1.016 \times \frac{5}{16}\frac{(\pi mkT)^{1/2}}{\pi\sigma^2} \tag{7}$$

and

$$\lambda_0 = 1.025 \times \frac{75k}{64m}\frac{(\pi mkT)^{1/2}}{\pi\sigma^2} \tag{8}$$

where k is the Boltzmann constant and T is the absolute temperature.

For a hard-sphere system, the Carnahan–Starling [10] formula

$$\chi = \frac{1 - b\rho/8}{(1 - b\rho/4)^3} \tag{9}$$

provides one with a quite accurate approximation for evaluating the transport coefficients. However, the interaction between real molecules is not of a rigid-sphere type and the above equation cannot be applied directly.

By attributing effective values to the covolume b, and the radial distribution function χ, deduced from compressibility isotherms for real gases, Eqs. (1) and (2) can be used for real gases. Enskog suggested that $b\rho\chi$ should be determined from compressibility measurements using the thermal pressure [8,9]

$$b\rho\chi = \frac{1}{\rho R}\left(\frac{\partial \mathrm{p}}{\partial T}\right)_{\rho} - 1 \tag{10}$$

where R is the gas constant, and p is the pressure. The modified Boltzmann equation introduced by Enskog will be reduced to the ordinary Boltzmann equation at low densities provided that

$$\lim_{\rho\to 0}(\chi) = 1 \tag{11}$$

Therefore, the problem is reduced to the specification of the thermal pressure, which, in turn, involves determining the isobaric expansivity and isothermal compressibility coefficients. These properties have been studied for n-alkanes by Flory et al. [11,12] and general correlations for a great variety of fluids have been obtained by Brostow et al. [13,14]. Amorós et al. [15,16] analyzed the behavior of the thermal pressure for several types of liquids and applied the modified Enskog theory to various fluids in the liquid range. Using the Peng–Robinson equation of state, Sheng et al. [17] calculated the transport properties of dense gases.

Here, we will try to obtain the parameters b and χ directly from an analytical equation of state based on statistical–mechanical perturbation theory, proposed by Song et al. [18, 19]. The equation of state reads as:

$$\frac{p}{\rho kT} = 1 + \frac{(B_2-\alpha)\rho}{1+0.22\Gamma\beta\rho} + \frac{\alpha\rho}{1-\Gamma\beta\rho} \tag{12}$$

where B_2 is the second virial coefficient, α is the contribution of the repulsive forces to the second virial coefficient, β is a temperature-dependent parameter analogous to the van der Waals covolume, and Γ is an adjustable parameter, equal to 0.454 for noble gases. All the temperature-dependent parameters can be determined knowing the intermolecular potential u, by the following three integrals:

$$B_2(T) = 2\pi\int_0^{\infty}\left(1-e^{-u/kT}\right)r^2\,\mathrm{d}r \tag{13}$$

$$\alpha(T) = 2\pi\int_0^{r_m}\left(1-e^{-u_0/kT}\right)r^2\,\mathrm{d}r \tag{14}$$

and

$$\beta(T) = 2\pi\int_0^{r_m}\left[1-\left(1+\frac{u_0}{kT}e^{-u_0/kT}\right)\right]r^2\,\mathrm{d}r \tag{15}$$

where $u_0(r)$ is the repulsive branch of $u(r)$ according to the Weeks–Chandler–Andersen [20] decomposition of $u(r)$. Substituting Eqs. (10) and (11) into Eq. (12) one may obtain:

$$\begin{aligned} b\chi = {} & \frac{B_2 + T\frac{\mathrm{d}B_2}{\mathrm{d}T} - \alpha - T\frac{\mathrm{d}\alpha}{\mathrm{d}T}}{1+0.22\Gamma\beta\rho} + \frac{\alpha + T\frac{\mathrm{d}\alpha}{\mathrm{d}T}}{1-\Gamma\beta\rho} \\ & + \frac{\Gamma\alpha\rho T\frac{\mathrm{d}\beta}{\mathrm{d}T}}{(1-\Gamma\beta\rho)^2} - \frac{0.22\Gamma\rho(B_2-\alpha)T\frac{\mathrm{d}\beta}{\mathrm{d}T}}{(1+0.22\Gamma\beta\rho)^2} \end{aligned} \tag{16}$$

with

$$b = B_2 + T\frac{\mathrm{d}B_2}{\mathrm{d}T} \tag{17}$$

Here, we will calculate Eqs. (1) and (2) for viscosity coefficient and thermal conductivity, respectively, from Eqs. (16) and (17).

3. Results and discussion

Our calculations are based on Eqs. (16) and (17) for χ and b, respectively. A simple Lennard–Jones (12-6) potential is used to evaluate the temperature-dependent parameters of the equation of state from Eqs. (13)–(15). The LJ (12-6) potential parameters together with the values of Γ for compounds studied in this work are listed in Table 1. For most of the compounds studied in this work the values of Γ are earlier reported by Ihm et al. [19]. For the others, we have obtained the best values of Γ from experimental $p\rho T$ data. Once the temperature-dependent parameters have been determined, the thermal conductivity and the viscosity can be calculated by knowing the values of λ_0 and η_0.

The values of λ_0 and η_0 differ from their corresponding values at ordinary pressures. Departures of the transport coefficients at atmospheric pressures from their values at zero-density limit have been discussed by Bich and Vogel [21] and it has been shown that the differences amount to several percent. The transport coefficients at zero-density limit can be obtained from the Chapman–Enskog theory [1], from the principle of corresponding states [22–25], or from the linear extrapolation of experimental low-density data. In this work the experimental viscosity data of Kr and the thermal conductivity data of Kr and Xe are taken from Van den Berg and Trappeniers [26] and Kestin et al. [27], respectively. The experimental viscosity data of Xe and the transport properties of the other noble gases, N_2,O_2,F_2,CO, CO_2,CH_4,C_2H_6 and C_3H_8 are taken from Vargaftik [28]. The transport prop-

Table 1
Lennard–Jones (12-6) potential parameters and the values of Γ for compounds studied

Substance	σ (nm)	ε/k (K)	Γ
He	0.2556	10.22	0.454
Ne	0.2780	34.9	0.454
Ar	0.3405	119.8	0.454
Kr	0.3827	164	0.454
Xe	0.4100	221.0	0.454
N_2	0.3677	95.14	0.418
O_2	0.3580	117.5	0.421
F_2	0.3334	108.75	0.419
CO	0.3656	96.2	0.420
CO_2	0.4124	218.8	0.359
CH_4	0.3808	152.0	0.418
CF_2Cl_2	0.5160	286.0	0.386
C_2H_6	0.4384	236.5	0.449
C_3H_8	0.5239	206.0	0.409
SF_6	0.5910	188.7	0.351

Table 2
Comparison of the calculated thermal conductivity of noble gases with experiment [28]

Substance	T (K)	$\Delta\rho$ (mol·m^{-3})	NP[a]	AAD[b] (%)	MD[c] (%)
He	270.00	45–11749	7	0.32	0.79
	300.00	40–10672	7	0.38	0.88
	400.00	32–8414	7	0.56	1.11
	500.00	24–6709	7	0.75	1.30
	600.00	21–5804	7	0.80	1.39
	700.00	18–4901	7	0.86	1.49
	800.00	16–4389	7	0.72	1.41
	900.00	14–3856	7	0.78	1.38
	1000.00	12–3519	7	0.64	1.01
	1100.00	10–3181	7	0.52	0.92
	1200.00	10–2952	7	0.56	0.92
	1300.00	9–2714	7	0.58	0.93
	1400.00	9–2545	7	0.49	0.82
	1500.00	8–2356	7	0.46	0.71
Ne	298.15	40–46367	18	0.89	2.45
	323.15	37–44532	18	1.05	2.19
	348.15	35–42841	18	1.48	2.41
Ar	150.00	81–5234	5	2.47	4.72
	200.00	60–4391	7	2.15	5.20
	300.00	40–6342	11	1.11	3.08
	400.00	30–8456	10	1.77	4.84
	500.00	24–11783	15	1.12	3.87
	600.00	20–10207	17	0.98	2.23
	700.00	17–8611	17	0.75	1.27
	800.00	15–7618	16	1.21	1.86
	900.00	13–6859	16	1.58	2.96
	1000.00	12–6246	13	1.72	3.37
	1100.00	11–5732	13	2.10	4.12
	1200.00	10–5293	13	2.17	4.06
	1300.00	9–4921	13	2.23	3.98
	1400.00	8–4547	13	2.34	3.67

[a] Number of points.

[b] Average absolute deviation.

[c] Maximum deviation.

Table 3
Comparison of the calculated thermal conductivities with experiment [28,29]

Substance	ΔT (K)	$\Delta\rho$ (mol·m^{-3})	NP	AAD (%)	MD (%)
N_2	100–1300	9–27233	560	1.38	6.81
O_2	200–1300	9–19201	325	1.89	7.35
CO_2	280–1300	9–25182	485	3.82	8.90
CH_4	213.15–573.15	21–22825	456	3.83	9.69
CF_2Cl_2	250–600	7–8162	281	3.46	8.21
C_2H_6	313.15–473.15	25–11110	71	4.11	10.27
SF_6	250–400	7–6243	78	3.57	9.21

Table 4
Comparison of the calculated viscosity coefficients for noble gases with experiment [26,28]

Substance	ΔT (K)	$\Delta\rho$ (mol·m^{-3})	NP	AAD (%)	MD (%)
He	77.35	161–45189	7	2.23	3.66
	157.15	83–27718	6	4.60	8.72
	194.65	63–23551	7	4.15	7.39
	273.15	44–26159	8	3.59	6.48
	373.15	32–20548	7	2.87	5.36
	473.15	25–16941	7	2.42	4.71
	773.15	16–7304	3	1.28	2.55
	1273.15	9–4531	3	0.71	1.47
Ne	50.00	241–6056	3	1.65	3.10
	100.00	124–12132	5	1.00	1.76
	160.00	75–7156	5	0.98	2.43
	200.00	64–5702	5	0.95	2.89
	298.15	43–33607	16	4.40	6.66
	323.15	42–34646	17	4.73	6.98
	348.15	41–35543	19	5.51	8.27
Ar	200.00	64–8441	11	0.46	1.85
	300.00	43–8389	16	0.50	2.63
	400.00	32–16072	23	0.84	3.28
	500.00	24–16606	26	0.41	0.71
	600.00	20–14407	26	0.89	1.81
	700.00	17–12781	26	1.25	2.99
	800.00	15–11388	26	1.33	3.41
	900.00	13–10321	26	1.53	4.35
	1000.00	12–9462	26	1.54	4.50
	1100.00	11–8744	26	1.55	4.67
	1200.00	10–8121	26	1.52	4.59
	1300.00	9–7589	26	1.50	4.52
Kr	298.15	122–3174	18	1.74	4.00
Xe	373.15	28–12947	4	2.99	7.51
	423.15	27–11641	5	2.79	7.81
	473.15	27–15718	6	3.31	7.45

Table 5
Comparison of the calculated viscosities with experiment [28,29]

Substance	ΔT (K)	$\Delta\rho$ (mol·m^{-3})	NP	AAD (%)	MD (%)
N_2	100–1300	9–26132	620	2.99	8.16
O_2	200–1300	9–8681	525	2.72	7.79
F_2	200–500	24–7234	63	2.83	6.97
CO	273.15–523.15	23–15870	60	2.49	6.28
CO_2	273.15–1273.15	9.5–3429	475	3.59	9.35
CH_4	200–500	24–17019	423	3.70	8.53
CF_2Cl_2	250–575	7–8162	245	3.49	7.33
C_2H_6	312.50–473.15	26–15148	105	4.26	9.09
C_3H_8	297.50–474.15	25–11544	92	2.69	7.25

erty data of CF_2Cl_2 and the thermal conductivity of SF_6 are taken from the recommended values by Heckenberger and Stephan [29]. For the sake of consistency we have used the same experimental data sources to obtain the zero-density parameters, λ_0 and η_0, at each temperature.

Following the above-mentioned procedure, the viscosity coefficient and the thermal conductivity of some simple fluids have been calculated with Eqs. (1) and (2) and are compared with the experimental values [26–29]. The average absolute deviation and the maximum deviation between our calculated thermal conductivities for He, Ne, and Ar over a wide range of temperatures and pressures are listed in Table 2. Also, the calculated thermal conductivity of N_2, O_2, CO_2, CH_4, CF_2Cl_2, C_2H_6, and SF_6 are compared with experiment [28,29] as average absolute deviation and maximum deviation in Table 3. The overall average absolute error for 2587 data points examined is 2.66% compared

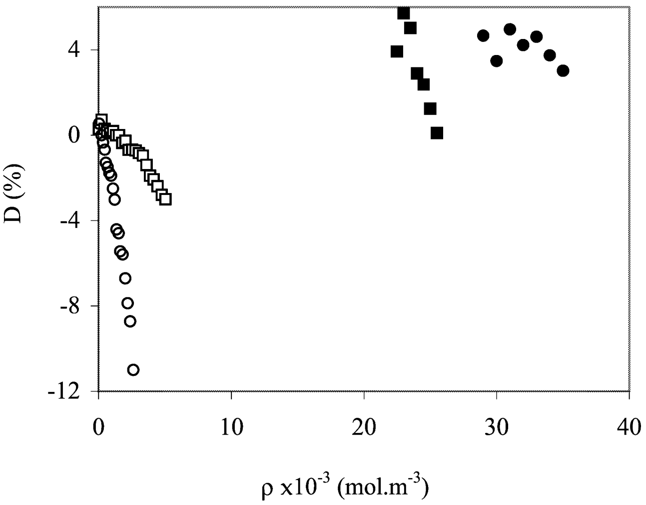

Fig. 1. Deviation plot for the thermal conductivity of Kr at 150.33 K (●) and 300.65 K (□) and that of Xe at 235.05 K (■) and 300.65 K (○) compared with experiment [27].

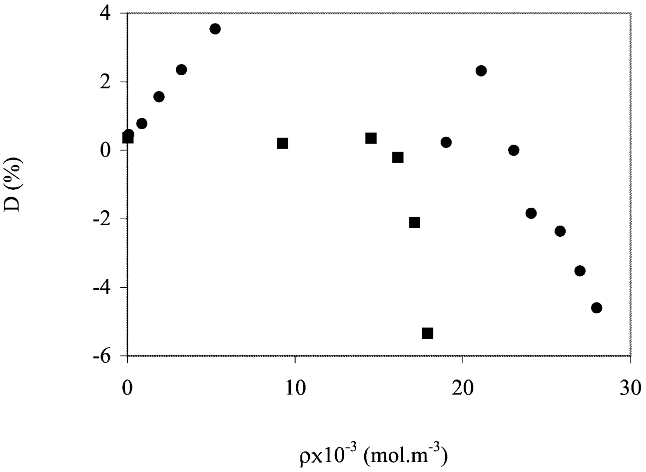

Fig. 2. Deviation plot for the viscosity coefficient of Ar at 150 K (●) and Xe at 323.15 K (■) compared with experiment [28].

to the tabulations of Vargaftik [28] and the recommended values of Heckenberger and Stephan [29], in the case of CF_2Cl_2 and SF_6. Also shown in Fig. 1 are the deviation percent between calculated and experimental values [27] of the thermal conductivity of Kr at 150.33 and 300.65 K and that of Xe at 235.05 and 300.65 K as a function of density. It is worth nothing that 300.65 K isotherm is near to the critical temperature of Xe, which is 289 K, and the thermal conductivity varies abruptly in the critical region [30]. This fact might be responsible for the departure of our predicted values from experimental data [27].

The average absolute deviation and the maximum deviation between calculated and experimental values [26,28,29] of viscosity for noble gases, N_2, O_2, F_2, CO, CO_2, CH_4, CF_2Cl_2, C_2H_6, and C_3H_8 are listed in Tables 4 and 5. The total average absolute deviation out of 3033 data points examined is 3.03%. The deviation percent between our calculated values of the viscosity coefficients of Ar at 150 K (the critical point of Ar) and Xe at 325.15 K compared with experiment [28] are also shown in Fig. 2. The higher deviations at low temperatures, Figs. 1 and 2, may partly be attributed to the larger inaccuracies in the predicted second virial coefficients, by Eq. (13), compared to the high temperatures.

Finally, we have combined the modified Enskog theory with an analytical equation of state based on the statistical–mechanical perturbation theory. The parameters of the equation of state can be determined from the intermolecular pair potential. Our proposed transport equation of state can predict the transport coefficients in a broad fluid range. Considering the overall good agreement with experimental values, it seems that our work meets demands for a simple estimation method for transport properties in terms of density and temperature. It is worth nothing that even the experimental values of the transport coefficients have some uncertainties, especially in the critical region.

Acknowledgements

The support of this work by Persian Gulf University Research Council under Grant No. 1-679-CsCHEM is gratefully acknowledged.

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