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The thermal conductivity of pure polyatomic gases at moderate pressure†

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Abstract—Based on the binary collisions of molecules, the kinetic theory of pure gases is valid for all pressures the higher the temperature increases. The mechanism of relaxation in the molecular internal energy exchange and the peculiar behaviour of polar gases are empirically taken into account through the reduced Planck constant (the de Boer parameter) to give an expression of the thermal conductivity.

INTRODUCTION

THE CHAPMAN–ENSKOG kinetic theory of dilute gases ignores the internal degrees of freedom of the molecules. Nevertheless it gives a good prediction of the viscosity of polyatomic gases through the Lennard–Jones (for non-polar gases) or Stockmayer (polar gases) interaction potential [1, 2]. This is possible because viscosity is insensitive to the effects of internal degrees of freedom. On the other hand, the thermal conductivity of polyatomic gases, non-polar or polar, is not described satisfactorily.

A monography with numerous references is given by Reid *et al.* [3]. Eucken [4] proposed the first attempt to patch up the basic theory and considered the ratio $\lambda M/\eta C_v$ where λ is the thermal conductivity, M the molar mass, η the viscosity coefficient and C_v the molar heat capacity at constant volume. That ratio, called the Eucken factor, is equal to 5/2 for monoatomic gases. To account for the effect of internal degrees of freedom, the molar heat capacity is split in two terms,

$$C_v = C_{\mathrm{tr}} + C_{\mathrm{int}},$$

where $C_{\mathrm{tr}} = (3/2)R$ is the translational heat capacity and $C_{\mathrm{int}} = C_v - C_{\mathrm{tr}}$ is the internal heat capacity. Then one decomposition of the same type is implemented on the Eucken factor

$$\frac{\lambda M}{\eta C_v} = f_{\mathrm{tr}}\left(\frac{C_{\mathrm{tr}}}{C_v}\right) + f_{\mathrm{int}}\left(\frac{C_{\mathrm{int}}}{C_v}\right). \tag{1}$$

Eucken proposed keeping $f_{\mathrm{tr}} = 5/2$ and $f_{\mathrm{int}} = 1$, whereby equation (1) reduces to

$$\left(\frac{\lambda M}{\eta C_v}\right)_{\mathrm{E}} = 1 + (9/4)R/C_v. \tag{2}$$

This relationship, named the *Eucken correlation*, which gives a very reasonable agreement with experiments at low temperature, permits one to calculate the 'low-temperature asymptotic conductivity' of non-polar gases when the values of M, η and C_v are known.

The molecules of the gas exist in various internal quantum states. It was noticed by some scientists that the transport of internal energy must occur by a diffusional mechanism and that it would be better to put into equation (1)

$$f_{\mathrm{tr}} = 5/2 \quad \text{and} \quad f_{\mathrm{int}} = Sc^{-1}$$

where the Schmidt number $Sc = \eta/\rho D$ is the ratio of the molecular momentum diffusivity (η/ρ) to the self-diffusion coefficient (D).

One way to verify this intuition by a theoretical approach is to consider each molecule with its excited internal energy state as one molecule of a separate chemical species and to assume that the transport of internal energy occurs by a mechanism suggested by chemical reaction into a mixture of chemical components [1, 5]. Then

$$\left(\frac{\lambda M}{\eta C_v}\right)_{\mathrm{mE}} = Sc^{-1} + \tfrac{3}{4}(5 - 2Sc^{-1})(R/C_v). \tag{3}$$

Using another method from the general formulas for the transport properties in Wang Chang and Uhlenbeck's form, Mason and Monchick [6] have obtained equation (3), which is named the *modified Eucken correlation.*

We notice that if $Sc = 1$, equation (3) is identical to equation (2). The numerical value of Sc^{-1} is near to 1.32 for gases and is nearly independent of temperature.

Equation (3) is valid when the interchange of rotational and translational energy is negligible, that is the collisions are quasi-elastic (the translational motion is independent of the internal states). Equation (3) is in very good accordance with experiments when the temperature is high for non-polar gases. Then it is possible to calculate the 'high-temperature asymptotic thermal conductivity' of non-polar gases.

The measured value of the thermal conductivity of

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† Dedicated to Professor Dr.-Ing. Dr.-Ing.e.h. Ulrich Grigull.

NOMENCLATURE

C_{int}	molar internal heat capacity
C_{tr}	molar translational heat capacity
C_v	molar heat capacity at constant volume ($C_v = C_{tr} + C_{int}$)
D_{int}	diffusion coefficient for internal energy
f_{int}	factor for internal heat capacity
f_p	corrective factor for polar gas
h	Planck constant
h^*	reduced Planck constant (or de Boer parameter)
k	Boltzmann constant
M	molar mass
m	mass of the molecule ($m = M/N$)
N	Avogadro constant
P	parachor
p	pressure
R	perfect gas constant
Sc	Schmidt number (for self-diffusion)
T	temperature.

Greek symbols

β	pressure coefficient of thermal conductivity
δ	polarity parameter
ε	energy parameter of the interaction potential
η	viscosity coefficient
λ	thermal conductivity
μ	dipole moment
π	constant in equations (6) and (7), $\pi = 3.1416$
π	molecular parachor ($\pi = P/N$)
ρ	density
σ	length parameter of the interaction potential
χ	physical property
$\Omega^{(2,2)*}$	reduced integral of collision for viscosity and thermal conductivity.

Superscripts

*	reduced relative to the (m, σ, ε, k) system
0	relative to the perfect gas.

Subscripts

E	Eucken correlation
int	internal
mE	modified Eucken correlation
tr	translational.

one non-polar gas lies between the two values obtainable by equations (2) and (3). Moreover, these equations indicate that the Eucken factor should always decrease with increasing temperature (with the growth of the heat capacity), but some experiments indicate an opposite behaviour in a certain range of temperatures the place of which, in the absolute temperature scale, depends on the studied gas.

In order to obtain theoretically a Eucken factor closer to reality, Mason and Monchick [6] consider that collisions are inelastic, with a relaxation time. They give relationships for f_{tr} and f_{int} showing that f_{tr} decreases and f_{int} increases when inelastic collisions occur. Neglecting the contribution of the vibrational energy, it is necessary to handle one new parameter: the collision number Z_{rot}, which is the number of collisions required to interchange a quantum of rotational energy with translational energy. It is difficult to find its value (which is temperature dependent) because experiments are tricky and no theory is well developed.

In all that has been written above, we have not considered a polar gas, the behaviour of which is markedly different. The thermal conductivity, anomalously low in relation to the viscosity, is due to a resonant exchange of rotational energy which is probable on grazing collision. This type of collision with exchange is equivalent to a head-on collision without exchange so far as the transport of the rotational quantum is concerned [6]. Thus, the diffusion coefficient for internal energy is smaller than the self-diffusion, and Brokav [7] gives an empirical correction

$$\rho D_{int}/\eta = Sc^{-1}/(1+\alpha) = 1.32/(1+\alpha)$$

with

$$\alpha = a(300/T)^n.$$

Values of a and n are given for highly polar gases: $0.016 < a < 0.93$, $1.6 < n < 2.1$.

The purpose of the present paper is to develop a simple empirical approach based on the principle of corresponding states. After van der Waals (1873) many scientists used this principle for various problems and especially to predict thermodynamic properties [1, 3, 8]. For the topic in question with transport properties, this principle may be applied with molecular parameters. Considering the case of noble gases, de Boer and Michels [9] introduced for the first time (1938) the dimensionless parameter which is the reduced Planck constant (sometimes called the de Boer parameter [1])

$$h^* = h/\sigma\sqrt{m\varepsilon}$$

where h is the Planck constant, m the molecular mass, and σ and ε the length and energy parameters in the molecular interaction potential. This parameter, which has a characteristic value for each gas (the greatest value being 2.64 for helium), appears as the

Broglie wavelength, divided by σ, of the relative motion of two molecules with relative kinetic energy ε [9, 11]. De Boer showed for the noble gases how evenly the reduced molecular volume and the internal energy at the absolute zero vary, and also the reduced Debye temperature when each property is plotted versus h^* [10].

Despite the ignorance of h^* in the application of the principle of the corresponding states, the experimental data concerning the viscosity of gases are well correlated [12, 13], but no such success is achieved for the thermal conductivity. The explanation lies in the fact that viscosity is insensitive to the effect of internal degrees of freedom. In some papers it is written erroneously that because h^* accounts for quantum effects, this parameter may be omitted for non-quantic gases: this depends on the type of property studied. Assuming that h^* has a central place in the internal energy of the molecule even for the 'classical' gases (non-quantic gases), we will point out that the thermal conductivity is a function of the reduced Planck constant.

THE PRINCIPLE OF CORRESPONDING STATES

The idea of corresponding states lies in the dimensional analysis. Any thermophysical property of one gas depends on two independent state variables (among three), the pressure p and the absolute temperature T, and also on a necessary and sufficient number of independent parameters which are characteristic of the chemical nature of the gas. In order to obtain dimensionless forms, the choice of the reference is free between the properties at the critical point or the intrinsic molecular properties. Using the last method, any transport property χ is a function of the molecular mass m, the two parameters of interaction σ and ε, the Boltzmann constant k, which is the molecular energy scale, and the Planck constant h, which is the scale of quantic mechanisms. For polar gases the dipole moment μ must be added. Then in the general case

$$\chi = \chi(p, T, m, \sigma, \varepsilon, k, h, \mu). \tag{4}$$

In this four-dimensional problem (length, mass, time, temperature), four parameters are kept for reference: m, σ, ε, k. The reduced form of equation (4) is

$$\chi^* = \chi^*(p^*, T^*, h^*, \mu^*), \tag{5}$$

where

$$p^* = p\sigma^3/\varepsilon \qquad T^* = kT/\varepsilon$$
$$h^* = h/\sigma(m\varepsilon)^{1/2} \quad \mu^* = \mu(\sigma^3\varepsilon)^{-1/2}.$$

When the attention is focussed on the viscosity or the thermal conductivity, χ^* is respectively identified with

$$\eta^* = \eta\sigma^2(m\varepsilon)^{-1/2} \quad \text{and} \quad \lambda^* = \lambda\sigma^2k^{-1}(m/\varepsilon)^{1/2}.$$

One notes here that it would be possible in equation (4) to replace h by a different parameter like the molecular parachor, which is the parachor P divided by the Avogadro constant N; then h^* would be replaced by $\pi^* = P\,(N^{0.25}\sigma^{2.5})^{-1}$ [14, 15]. Each of the two cases describe the state of the gas in a coherent (uniqueness) manner; there exists necessarily a relationship between h^* and π^* (through μ^* for polar gases). However, h^* is preferred here because it is closer to the physical model of internal energy.

For dilute gases, the reduced pressure p^* is cancelled in equation (5) and considering monoatomic gases, the Chapman–Enskog theory leads to

$$\eta^* = \frac{5}{16\sqrt{\pi}}\frac{\sqrt{T^*}}{\Omega^{(2,2)*}} \tag{6}$$

and

$$\lambda^* = \frac{75}{64\sqrt{\pi}}\frac{\sqrt{T^*}}{\Omega^{(2,2)*}} \tag{7}$$

where $\Omega^{(2,2)*}$ is a reduced collision integral and is a function of T^*.

For non-polyatomic gases, equation (6) is applicable without alteration. Because the viscosity is insensitive to the internal degrees of freedom, h^* is not active. For polar gases we must consider in equation (6) the collision integral $\Omega^{(2,2)*}$ (T^*, μ^*). In place of μ^*, Monchick and Mason [2] have given numerical tables of $\Omega^{(2,2)*}$ (T, δ) where $\delta = (\mu^*)^2/2$. Brokav [7] has proposed an analytical expression which is easy to handle.

Applying equation (5) to the Eucken factor, which is dimensionless, one obtains the general form for dilute gases

$$\frac{\lambda M}{\eta C_v} = f(T^*, h^*, \delta). \tag{8}$$

Before further discussion about the thermal conductivity, we have to examine the question: it is generally admitted that the formulae of the kinetic theory are valid for gases at atmospheric pressure, but what is the upper limit of pressure?

THE KINETIC THEORY AT MODERATE PRESSURE

The Chapman–Enskog theory of gases is based on binary collisions of molecules. According to the assumption of a dilute gas, the viscosity is pressure-independent at any temperature. In order to evaluate the higher limit of pressure compatible with binary collision, the paper of Trappeniers *et al.* [13] is useful. Keeping numerous precise data on the viscosity of noble gases up to high densities, these authors give an empirical series expansion in terms of the reduced density ρ^* and reduced temperature T^*. One can use this relationship for low density, noting that for dilute gases $\rho^* = p^*/T^*$. It is possible to estimate the upper pressure limit so that the viscosity increase is less than

0.5% (that is, the level of uncertainty of the best data). This requirement is fulfilled if

$$p < 6.4 \times 10^4 \frac{T}{\sigma^3} \Omega^{(2,2)*} \quad \text{for} \quad 0.45 < T^* < 15 \qquad (9)$$

with σ in Å (0.1 nm), T in K and p in Pa (N m^{-2}).

Taking into account the insensitivity of viscosity to the internal degrees of freedom, this condition is certainly valid for non-polar gases. This means that for nitrogen at 300 K, equation (6) is applicable up to 3.4 bars; at 1000 K, the upper limit is 9 bars.

Based on the empirical relationship given for polar gases at low density [16], several checks show that (9) is still valid in spite of the peculiar behaviour of these gases. For example, the viscosity of ammonia at 300 K can be calculated by the kinetic theory for pressure up to 6.5 bars.

It can be assumed that the conclusion obtained with viscosity is valid for thermal conductivity of monoatomic gases. For polyatomic gases, the variation in thermal conductivity with pressure is stronger due to the variation in internal heat capacity. The analysis of the problem is complicated by the fact that the accuracy of data or conductivity is about ±1% in the better experiments. We will see later that it is possible to calculate the thermal conductivity inside the pressure range defined by (9) where the binary collisions are predominant.

One notices that in practice the range of pressure where the theory is applicable, increases with temperature.

AN EMPIRICAL FORMULATION

Touloukian *et al.* [17–19] have given precious information covering pure gases. They have reviewed and analysed all the data and 'recommended reference values' are presented as the most probable. Based on that work we are led to conclude that the Eucken factor is known with a margin of error estimated at ±6% in usual cases. The most important uncertainty weighs on thermal conductivity and heat capacity values and this explains the fact why some Eucken factors, become greater than the values given by equation (3) when the temperature increases.

Consequently we will neglect the translational/rotational interchange in assuming that: (i) f_{tr} is equal to 5.2 as in equations (2) and (3); (ii) the variation of the relaxation time with temperature is solely included in f_{int}

$$f_{int} = 1 + (Sc^{-1} - 1) \exp(-10h^*/\sqrt{T^*}). \qquad (10)$$

Through this expression f_{int} varies from unity at weak temperature (no internal interchange) up to Sc^{-1} at high temperature (very fast interchange).

The de Boer parameter is practically

$$h^* = 43.764/\sigma\sqrt{M(\varepsilon/k)} \qquad (11)$$

with σ in Å, ε/k in K and M in g mol^{-1}.

As was noted in the Introduction, the thermal conductivities of polar gases are low in relation to the viscosity. In order to take this fact into account, we introduce a corrective factor

$$f_p = \exp(-11h^{*2}\delta/T^*) \qquad (12)$$

which multiplies f_{int} so that equation (3) becomes

$$\frac{\lambda M}{\eta C_v} = f_p f_{int} + \frac{3}{4}\frac{R}{C_v}(5 - 2f_p f_{int}), \qquad (13)$$

this relationship being associated with equations (10) and (12).

Table 1 presents some cases selected from Table 10-2 of ref. [3], in order to allow an easy comparison with other types of empirical evaluation considered in that reference. The influence of the chosen property values is noticeable through recommended values added in the table. We can conclude that the calculated Eucken factor is within the margin of experimental uncertainty. The constants in equations (10) and (12) were not selected by a severe method of optimization and they can be improved. Nevertheless, the deviations are still statistically equal to those of the more accurate method recommended by Reid *et al.* [3].

Based on this analysis, the thermal conductivity of gases is evaluated inside the domain where the kinetic theory is applicable (condition (9))

$$\lambda = \frac{R}{M}\left[\frac{15}{4} + f_p f_{int}\left(\frac{C_v}{R} - \frac{3}{2}\right)\right]\eta. \qquad (14)$$

At weak density, C_v is equal to C_v^0 for perfect gases and $C_v^0/R = (C_p^0/R) - 1$. At moderate pressure we take into account the influence of pressure.

THE VARIATION OF CONDUCTIVITY WITH PRESSURE

It is possible to use equation (14) when the pressure is greater than one atmosphere. The increase of thermal conductivity is due to the increase of the heat capacity with pressure. At constant temperature, this increase can be characterized by the pressure coefficient

$$\beta = \frac{1}{\lambda_0}\frac{d\lambda}{dp} = \frac{R}{M}\frac{\eta}{\lambda_0} f_p f_{int} \frac{d}{dp}\left(\frac{C_v}{R}\right)$$

where λ_0 is the conductivity at weak density. Then, for a moderate pressure, the evaluation of λ and β requires the use of an equation of state in order to find the C_v value or its derivative. A survey of numerical tables [20] shows that for a given gas at constant temperature $d/dp(C_v/R)$ is positive and decreases when the fixed temperature grows. This conclusion is compatible with the observations on β made by Vines and Bennett [21]. Comparison between calculated and experimental values of β is satisfying if it is noticed that the experimental approach of β values can be in error by at least ±50% [21]. The coefficient β varies greatly from one gas to another and is about 0.5% per

Table 1. Comparison between calculated and experimental values of the Eucken factor of a pure gas at 1 bar (see Table 10-2 in ref. [3])

						Percent error†		
Component	T (K)	λ (10^{-3} W m^{-1} K^{-1})	η (10^{-6} N s m^{-2})	C_v (J mol^{-1} K^{-1})	Eucken factor	Eucken	Mod. Euck.	Present work
Acetylene	198	11.8	7.01	26.8	1.64	3.8	14	5.8
	273	18.7	9.55	33.7	1.51	2.8	16	6.6
		18.5‡	9.45	33.5	1.52			5.9
	373	29.8	12.61	40.4	1.52	−4.0	10	1.0
		29.7	12.65	40.4	1.51			1.6
Benzene	353	14.6	9.0	90.4	1.40	−14	5.8	−1.8
		14.67	8.99	90.56	1.407			−2.2
	433	22.6	10.95	114	1.41	−18	2.5	−5.0
		23.59	11.0	120.8	1.38			−1.9
n-Butane	273	13.5	6.88	84.6	1.35	−9.4	11	0
		13.5	6.93	83.3	1.36			0.7
	373	24.6	9.45	110	1.38	−15	5.7	−4.3
		23.4	9.33	109.3	1.33			−0.7
n-Hexane	373	20.1	7.90	163	1.34	−17	4.8	−2.1
	433	27.2	9.20	186	1.37	−20	2.1	−4.4
		27.16	9.42	187.5	1.32			−0.7
Acetone	353	15.7	9.0	77.9	1.30	−4.7	16	2.9
		15.74	8.97	77.3	1.32			1.4
	393	19.4	10.0	84.2	1.34	−8.7	12	−0.3
	457	24.7	11.45	96.1	1.30	−8.4	13	1.5
		25.9	11.6	96	1.35			−1.9
Ammonia	213	16.5	7.32	25.4	1.51	15	26	2.7
		16.47	7.306	25.4	1.51			2.7
	273	21.9	9.06	26.7	1.54	10	21	0.5
Sulphur dioxide	273	8.29	11.7	30.6	1.48	8.6	21	−3.2

† Percent error = [(calc. − exp.)/exp.] × 100; $Sc^{-1} = 1.32$.

‡ Recommended values are underlined.

bar, depending on the temperature and the chemical nature.

Equation (14) is then usable for moderate pressure delimited by condition (9), that is in the range of pressure where the viscosity coefficient is practically constant (chiefly binary collisions). The latter property is obtained by equation (6), which is written here in the classical form

$$\eta = 2.6693 \times 10^{-6} \frac{\sqrt{MT}}{\sigma^2 \Omega^{(2,2)*}} \qquad (15)$$

where η is in Pl (or N s m^{-2}), M in g mol^{-1}, T in K and σ in Å (or 0.1 nm).

The margin of error on the calculated conductivity value (equation (14)) is $\pm 2\%$.

CONCLUSION

Through the analysis of the variation of viscosity with pressure at constant temperature, a range of pressure (reaching several atmospheres) is defined in which the kinetic theory is applicable. When recommended values replace rough data of physical properties the variations of Eucken factor and temperature are opposite. Based on these two critical comments, the intensity of internal energy exchange is empirically associated to temperature, reduced Planck constant and reduced dipole moment in order to obtain an improved expression on the thermal conductivity of polyatomic gases.

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APPENDIX

	M (g mol^{-1})	σ (Å)	ε/k (K)	h^*	δ
Acetylene	26.04	4.033	231.3	0.1398	0
Benzene	78.11	5.349	412.3	0.04559	0
n-Butane	58.12	4.687	531.4	0.05313	0
n-Hexane	86.18	5.949	399.3	0.03966	0
Acetone	58.08	4.50	549	0.05446	0.67
Ammonia	17.03	2.90	464	0.1698	0.69
Sulphur dioxide	64.06	3.49	343	0.08459	0.21

LA CONDUCTIVITE THERMIQUE DES GAZ PURS POLYATOMIQUES AUX PRESSIONS MODEREES

Résumé—Basée sur l'hypothèse de collision binaire des molécules, la théorie cinétique des gaz purs est valable jusqu'à des pressions d'autant plus élevées que la température croît. Le mécanisme de relaxation dans l'interchange des énergies internes moléculaires et le comportement particulier des gaz polaires sont pris en compte empiriquement à l'aide de la constante de Planck réduite (ou paramètre de De Boer) pour exprimer la conductivité thermique.

DIE WÄRMELEITFÄHIGKEIT REINER MEHRATOMIGER GASE BEI MITTLEREN DRÜCKEN

Zusammenfassung—Die kinetische Theorie reiner Gase, die auf binären Kollisionen von Molekülen beruht, gilt für um so höhere Drücke je höher die Temperatur ist. Der Mechanismus der Relaxation beim molekularen Austausch innerer Energie und das spezielle Verhalten polarer Gase werden empirisch mit Hilfe der normierten Planck'schen Konstanten (dem de Boer Parameter) berücksichtigt. Damit ergibt sich ein Ausdruck für die Wärmeleitfähigkeit.

ТЕПЛОПРОВОДНОСТЬ ЧИСТЫХ МНОГОАТОМНЫХ ГАЗОВ ПРИ УМЕРЕННОМ ДАВЛЕНИИ

Аннотация—Кинетическая теория чистых газов, основанная на парных соударениях молекул, справедлива до давлений, соответствующих самым высоким температурам. В выражении для теплопроводности механизм релаксации при обмене внутренней энергии молекул и поведение полярных газов эмпирически учитываются с помощью приведенной константы Планка (параметра де Бура).