The Thermal Conductivity of Nitrogen and Carbon Monoxide in the Limit of Zero Density

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The paper presents accurate representations for the thermal conductivity of the diatomic gases nitrogen and carbon monoxide in the limit of zero density. These gases were studied because they have nearly the same molecular mass and viscosities. In contrast, the new analysis confirms that the thermal conductivities of the two gases differ remarkably, especially at low temperatures. The theoretically-based correlations provided are valid for the temperature range 220–2100 K and have associated uncertainties of $\pm$ 1% between 300 and about 500 K, rising to $\pm$ 2.5% at the low- and high-temperature extremes. A comparison with some empirical and semiempirical correlations is given.

Key words: carbon monoxide; nitrogen; polyatomic gases; thermal conductivity of gases; transport properties.

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1. Introduction

Much of the analytic work connected with the efficient design or optimization of industrial processes has been taken over by complex computer packages. To be effective, such computer codes must include powerful databases for the representation of the thermophysical properties of fluids that can produce accurate and reliable numerical data over the widest possible range of thermodynamic states. So far as transport properties are concerned, it is conventional, for these purposes, to express the properties in the form

$$X(\rho,T) = X_0(T) + \Delta X(\rho,T) + \Delta X_c(\rho,T). \quad (1)$$

Here the term $X_0(T)$ represents the transport property—e.g., viscosity or thermal conductivity—in the limit of zero density. The term $\Delta X(\rho,T)$ represents the excess property and $\Delta X_c(\rho,T)$ the critical enhancement. From this formula it is obvious that the starting point for the correlation of the transport properties of fluids over a wide range of thermodynamic states must always be an analysis of the behavior of the properties in the limit of zero density. In this paper we extend earlier studies of the viscosity of nitrogen and oxygen[1], as well as of carbon dioxide, methane, and sulfur hexafluoride[2] to the thermal conductivity of nitrogen and carbon monoxide in the same limit.

The most recent critical review of the thermal conductivity of nitrogen in the limit of zero density, based on a kinetic theory analysis, was published some 15 years ago.[3] For carbon monoxide no correlations of this type are available in the literature. Since the publication of the earlier analysis for N_2, several new measurements using improved experimental techniques have been made and they are generally viewed with a higher level of confidence than the earlier results. Furthermore, there have been some notable theoretical advances in the treatment of the thermal conductivity of polyatomic gases,[4-6] although the subject has not yet progressed to the stage characteristic of that for the monatomic gases.[6] Thus, whereas a formal kinetic theory exists for the thermal conductivity of both monatomic and polyatomic gases, it is only possible to perform exact and routine calculations from an intermolecular pair potential for the former. This is because the anisotropic potentials characteristic of the interactions of polyatomic molecules, as well as the internal energy such molecules possess, complicate the dynamics of binary collisions sufficiently to place the calculation beyond the range of even the fastest of modern computers. For this reason the development of a correlation scheme for the thermal conductivity of polyatomic gases can derive guidance from theory but must, to a great extent, rely upon high precision experimental measurements.

The aim of this paper is the presentation of a correlation of a critically-evaluated set of experimental data on the thermal conductivity of nitrogen and carbon monoxide which is consistent with the available kinetic theory, and both accurate and simple to use. The ultimate aim of such work should be to cover the temperature range from the triple point, (T_t, P') to above the critical point (T_c, P_c). For nitrogen T_t = 63.15 K, P_t = 0.012 53 MPa, T_c = 126.20 K, and P_c = 3.4000 MPa, whereas for carbon monoxide T_t = 68.13 K, P_t = 0.0154 MPa, T_c = 132.85 K, and P_c = 3.494 MPa.

In the present report the representation is confined to the zero density limit and to temperatures above 220 K for reasons discussed later.

2. Experimental Data

From a preliminary analysis of the entire set of published thermal conductivity data for nitrogen and carbon monoxide, it is apparent that there are substantial discrepancies between various results. Since it is extremely difficult to decide on the accuracy of reported data solely on the basis of the available literature we have also employed two complementary methods of assessing, in particular, the older experimental data. First, we use a comparison with new experimental data acquired with improved, modern equipment of proven accuracy. Secondly, we attempt to establish confidence in experimental data by recourse to the available kinetic theory. Thus, we commence our analysis by dividing the experimental data into two categories.[1,2,7]

(i) Primary data

These are the results of measurements made with a primary apparatus of high precision for which a complete working equation and a detailed knowledge of all corrections is available. Strictly, for the thermal conductivity this means that only measurements made with the transient hot-wire technique are included. However, because this would reduce the temperature range studied unduly, we have also included data in the primary data set those results obtained by different techniques, which prove consistent with the transient hot-wire data and theoretical results.

(ii) Secondary data

These are the results of measurements which are of inferior accuracy to primary data. This inferior accuracy may arise from operation at extreme conditions or from incomplete characterization of the apparatus.

Among the secondary data two trends can be observed. First, there is a group for which the absolute thermal conductivity is significantly in error, but for which the temperature function is consistent with that of the present correlation. Second, there are data sets for which both the absolute value and the temperature dependence are markedly different.

For nitrogen as well as carbon monoxide, we have carried out a survey of the available data and assigned them to one of the above-mentioned categories. Only primary data were used in the formulation of the final correlations. A significance is attached to each datum determined by our estimate of the uncertainty. In the following sections the data selected as primary for each system are detailed, together with our estimate of their uncertainty, a summary of which is included in Table 1.

In most cases, the thermal conductivity data we employ have been acquired through measurements near or below atmospheric pressure or through measurements as a function of density at high pressures. In the latter case, we have made use of the results of the original statistical analyses of

Table 1.   Primary experimental data for the thermal conductivity of nitrogen and carbon monoxide

Reference	Technique	Temperature Range (K)	Ascribed Uncertainty (%)
Nitrogen			
Haran et al.[9]	THW	308–430	± 0.5
Johns et al.[10,11]*	THW	345–470	± 0.5
Clifford et al.[13]	THW	301	± 0.5
Haarman[14]	THW	328–468	± 1.0
Dijkema[16]	HW	300	± 1.0
Guildner[17]	HW	282, 348	± 1.0
Tufeu et al.[18]	CC	298	± 2.5
Le Neindre[19]	CC	300–942	± 2.5
Carbon Monoxide			
Assael et al.[8]	THW	308	± 0.5
Haran et al.[29]	THW	337–430	± 0.5
Imaishi et al.[31]	THW	301	± 0.5
Dijkema et al.[16]	HW	300	± 1.0
Johnston et al.[32]	HW	87–377	± 3.0

THW = Transient hot wire

HW = Hot wire

CC = Concentric cylinders

*Following a private communication from the authors, the data of Ref. 12 have not been included because they are thought to have been in error.

the data as a function of density in order to obtain values appropriate to the zero density limit.

2.1. Thermal conductivity data

2.1.1. Nitrogen

The most accurate of the thermal conductivity measurements have been carried out with transient hot-wire instruments at Imperial College,[8,9] NEL, Glasgow,[10,11,12] and Brown University.[13] The measurements at Imperial College extend over the temperature range 308–430 K, those at NEL over the range 345–470 K, whereas, at Brown, one point at 300.65 K has been measured. The uncertainty ascribed here to the zero density values derived from these measurements is $\pm 0.5\%$.

Using the criteria of the previous section, the remaining primary data are the results of measurements, generally performed using different methods, which are of inferior accuracy to those from the transient hot-wire instruments mentioned above. The results of Haarman[14] carried out in a transient hot-wire apparatus, are estimated to have an uncertainty of $\pm 1.0\%$. This exceeds the author's own estimate of $\pm 0.3\%$ but is consistent with the observed behavior of the results of the same apparatus for monatomic gases.[15] The ascribed uncertainty for the results of Dijkema et al.[16] and Guildner[17] is also $\pm 1.0\%$, whereas, the data of Le Neindre et al.[18,19] are estimated to have an uncertainty of $\pm 2.5\%$. The latter results have a significantly greater uncertainty but they extend the temperature range of the correlation substantially beyond that covered by the transient hot-wire instruments. Within the category of secondary data we have not attempted to be comprehensive and have included only selected data sets. This is because the general trends for the literature before 1972 can be easily deduced from our comparison with the correlation of Hanley and Ely,[3] and the deviation plots given in Ref. 3, which were based on these early data. Therefore, we compare the present correlation only with the results of work that was carried out since 1972 and with some more recent correlations.[20–30]

2.1.2. Carbon monoxide

Again we include the data from Imperial College[8,9] and Brown University[31] in the primary data set. The former extend over the temperature range 308–430 K: the ascribed uncertainty is $\pm 0.5\%$. There is one point at 300.65 K from Brown with the same ascribed accuracy.

The remaining data in the primary data set are those of Dijkema et al.[16] (300 K, $\pm 1.0\%$) and Johnston and Grilly[32] (296–377 K, $\pm 3\%$). The data of Saxena[33] have been classified as secondary data. For comparison, we also include the recent correlation of Matsunaga et al.[29]

3. Kinetic Theory Analysis

As already mentioned it became apparent from a preliminary analysis of the available data that there are substantial discrepancies between them, especially at the extremes of the temperature region. We have, therefore, attempted to establish confidence in experimental data by recourse to the kinetic theory of gases. The details of such an analysis have been given for carbon dioxide elsewhere[34] so that we here present only a condensed version.

3.1. Theoretical Results

3.1.1. Viehland–Mason–Sandler Formalism

According to the kinetic theory the thermal conductivity of a polyatomic gas in the limit of zero density may be written as a sum of two contributions

$$\lambda = \lambda_{tr} + \lambda_{int}, \tag{2}$$

in which

$$\lambda_{tr} = \frac{5k^2T}{2m\langle v\rangle_0}\left\{\frac{\mathfrak{S}(1001) + r\mathfrak{S}(^{1010}_{1001})}{\mathfrak{S}(1010)\mathfrak{S}(1001) - \mathfrak{S}^2(^{1010}_{1001})}\right\} \tag{3}$$

and

$$\lambda_{int} = \frac{5k^2T}{2m\langle v\rangle_0}\left\{\frac{r\mathfrak{S}(^{1010}_{1001}) + r^2\mathfrak{S}(1010)}{\mathfrak{S}(1010)\mathfrak{S}(1001) - \mathfrak{S}^2(^{1010}_{1001})}\right\}S. \tag{4}$$

The quantities $\mathfrak{S}(^{pqrs}_{p'q'r's'})$ are effective cross sections that incorporate all of the information about binary molecular collisions and, hence, the intermolecular pair potential. [If $pqrs \equiv p'q'r's'$ we adopt the usual notation $\mathfrak{S}(^{pqrs}_{p'q'r's'}) = \mathfrak{S}(pqrs)$.]

The additional symbols introduced in Eqs. (2) to (4) are

$$\langle v\rangle_0 = 4(kT/\pi m)^{1/2}, \tag{5}$$

$$r = (2c_{int}/5k)^{1/2} \tag{6}$$

and

$$S \approx 1 - (5/3)(1 + \lambda_{tr}/\lambda_{int})(\Delta\lambda_\parallel/\lambda)_{sat}. \tag{7}$$

Here, c_{int} is the internal heat capacity of the gas, m the molecular mass of the gas and S is a correction factor to the semiclassical Wang Chang–Uhlenbeck kinetic theory that accounts for spin polarization effects.[4]

There exist a number of exact relationships between different cross sections.[36] Here we make use of the results

$$\mathfrak{S}(^{1010}_{1001}) = (5r/6)\mathfrak{S}(0001), \tag{8}$$

$$\mathfrak{S}(1010) = (2/3)\mathfrak{S}(2000) + (25r^2/18)\mathfrak{S}(0001) \tag{9}$$

and

$$\mathfrak{S}(0010) = (5r^2/3)\mathfrak{S}(0001). \tag{10}$$

The cross section $\mathfrak{S}(2000)$ is related to the zero density viscosity by the equation

$$\eta_0(T) = \frac{kT}{\langle v\rangle_0}\mathfrak{S}(2000)^{-1}, \tag{11}$$

whereas, for the (hypothetical) coefficient of self-diffusion we have

$$D = \frac{kT}{nm\langle v\rangle_0}\mathfrak{S}(1000)^{-1} \tag{12}$$

and for the collision number for internal energy relaxation[37]

$$\xi_{int} = \frac{4kT}{\pi\eta\langle v\rangle_0}\mathfrak{S}(0001)^{-1}. \tag{13}$$

Finally, the so-called diffusion coefficient for internal ener-

$_{\text{F }}$' has been defined as [Eq. (16) in Ref. 4 is missing a factor k/m]

$$D_{\text{int}} = \frac{kT}{nm\langle v\rangle_0}\left[\mathfrak{S}(1001) - \left(\frac{1}{2}\right)\mathfrak{S}(0001)\right]^{-1}. \quad (14)$$

From Eqs. (2), (4), and (8)–(10), it becomes obvious that the thermal conductivity λ can be related to just three effective cross sections $\mathfrak{S}(2000)$, $\mathfrak{S}(0001)$, and $\mathfrak{S}(1001)$. It is also evident that, if measurements of the viscosity, total thermal conductivity, ξ_{int} and $(\Delta\lambda_{\|}/\lambda)_{\text{sat}}$ are available, or alternatively, if the viscosity, thermal conductivity, its translational part, λ_{tr}, and $(\Delta\lambda_{\|}/\lambda)_{\text{sat}}$ are known, then the foregoing equations can be used to evaluate all of the cross sections mentioned explicitly and consistently. Fortunately, for nitrogen and carbon monoxide all of the required information is available and this provides a means of assessing the consistency of the experimental thermal conductivity data.

In this assessment we make use of the further kinetic theory result that

$$\frac{D_{\text{int}}}{D} = \frac{5D_{\text{int}}nm}{6\eta A^*}, \quad (15)$$

where A^* is a cross section ratio[6] very weakly dependent on the pair potential between molecules. This last result can be put to good use because the theoretical results of Moraal, McCourt, and Snider[36,38] demonstrate that for diatomic molecules at high temperatures

$$\mathfrak{S}(1001) \to \mathfrak{S}(1000) + (7c_{\text{int}}/6k)\mathfrak{S}(0001), \quad (16)$$

(In the relevant relation of Table II in Ref. 38 the factor k/c_{int} must be cancelled) and, therefore, that

$$\frac{D_{\text{int}}}{D} = \frac{\mathfrak{S}(1000)}{\mathfrak{S}(1000) + \left[\left(\dfrac{7c_{\text{int}}}{6k}\right) - \dfrac{1}{2}\right]\mathfrak{S}(0001)}, \quad (17)$$

so that according to theory, D_{int}/D approaches unity from below in the high-temperature limit, since $\mathfrak{S}(0001) \to 0$.

3.2. Formalism of Thijsse *et al.*

Thijsse *et al.*[5] have found that the thermal conductivity of a polyatomic gas may also be written in the form

$$\lambda_0 = \frac{5k^2T(1+r^2)}{2m\langle v\rangle_0\mathfrak{S}(10E)}\left[1 - \frac{\mathfrak{S}^2(^{10E}_{10D})}{\mathfrak{S}(10E)\mathfrak{S}(10D)}\right]^{-1}, \quad (18)$$

where the new effective cross-sections $\mathfrak{S}(10E)$, $\mathfrak{S}(10D)$, and $\mathfrak{S}(^{10E}_{10D})$ are related to those introduced earlier.[35]

By making use of these exact relationships it has been found[5,35] that for N_2 and CO, among other gases examined,

$$\mathfrak{S}^2(^{10E}_{10D}) \ll \mathfrak{S}(10E)\mathfrak{S}(10D) \quad (19)$$

so that the simpler formula

$$\lambda_0 = \frac{5k^2T}{2m\langle v\rangle_0}\frac{(1+r^2)}{\mathfrak{S}_T(10E)} \quad (20)$$

departs[35] from the full form of Eq. (18) by no more than 0.9% in the temperature range above 300 K. In Eq. (20) we introduce the subscript T in order to distinguish this cross section from $\mathfrak{S}(10E)$ to which it is merely an approximation.

We shall see later that for nitrogen and carbon monoxide Eq. (20) provides an extremely concise means of representing the thermal conductivity data.

4. Applications of the Kinetic Theory Analysis

In order to carry out a kinetic theory analysis of thermal conductivity, it is necessary to have information about the temperature dependence of the viscosity in the limit of zero density, of the ideal-gas heat capacity, the collision number of internal energy relaxation ζ_{int}, and on the fractional change of the thermal conductivity in a magnetic field at saturation $(\Delta\lambda_{\|}/\lambda)_{\text{sat}}$. For nitrogen as well as for carbon monoxide, this information is available from the literature and the sources are summarized in Table 2.

Table 2. Sources of auxiliary data for the kinetic theory

analysis of the thermal conductivity

| Property | Nitrogen | | Carbon Monoxide | |
	Reference	Temperature Range (K)	Reference	Temperature Range (K)
Viscosity	[1, 40]	120–1200	[1, 40]	120–2100
C_p^0	[43]	120–2500	[49, 50]	68–2100
$\left[\begin{matrix}\mathfrak{S}(0001)\\\zeta_{\text{coll}}\end{matrix}\right]$	[42]	120–2100	[42]	120–2100
$(\Delta\lambda_{\|}/\lambda)_{\text{sat}}$	[24, 45]	temperature independent	[24, 45]	temperature independent

Table 3. Coefficients of Eq. (25) for the representation of the
effective cross-sections of nitrogen and carbon monoxide

	$\mathfrak{S}^*(2000)$	$\mathfrak{S}^*(0001)$	$\mathfrak{S}^*(1001)$	D_{int}/D
	Nitrogen			
	$120 \leq T \leq 2100$	$300 \leq T \leq 2100$	$300 \leq T \leq 2100$	$220 \leq T \leq 430$
a_0	4.812904	−0.28729	3.609527	−0.1187821
a_1	−0.472230	−1.01857	−14.518969	0.3864551
a_2	0.0878508	0.429487	16.820440	−0.6802974
a_3	0.0107001	−0.179227	−10.529320	0.5410572
a_4	−0.00518589	0.050901	3.475500	−0.1494064
a_5	0.0	−0.0060826	−0.575151	0.0
a_6	0.0	0.0	0.0375182	0.0
	Carbon Monoxide			
	$120 \leq T \leq 2100$	$300 \leq T \leq 2100$	$300 \leq T \leq 2100$	$220 \leq T \leq 485$
a_0	4.812904	−0.808156	−0.032112	−0.2830483
a_1	−0.472230	−0.6405754	0.0	0.0898495
a_2	0.0878508	0.0	−2.296496	−0.1522011
a_3	0.0107001	0.0	2.256431	0.3541263
a_4	−0.00518589	−0.1498986	−0.995782	−0.1401692
a_5	0.0	0.08018675	0.2149422	0.0
a_6	0.0	−0.01125297	−0.0184596	0.0

4.1. Nitrogen

The viscosity of nitrogen in the limit of zero density has been examined by Cole and Wakeham.[1] They provided a representation of the viscosity in the temperature range 110–2100 K with an associated uncertainty of $\pm 0.3\%$ near room temperature rising to $\pm 2\%$ at either extreme of the temperature range.

Since the preparation of that representation, a new series of precise measurements with the oscillating-disk viscometer in Rostock (GDR)[39,40] has been performed. These data, and an additional set of Timrot et al.,[41] have been added to the primary data set and a new correlation prepared.[40] In terms of the reduced effective collision cross section $\mathfrak{S}^*(2000)$, it is

$$\eta_0(T) = \frac{kT}{\langle v\rangle_0}[\pi\sigma^2\mathfrak{S}^*(2000)]^{-1}, \tag{21}$$

with

$$\mathfrak{S}^*(2000) = \mathfrak{S}(2000)/\pi\sigma^2, \tag{22}$$

$$T^* = kT/\epsilon \tag{23}$$

and

$$\epsilon/k = 104.2 \ \text{K}, \quad \sigma = 0.3632 \ \text{nm}. \tag{24}$$

The temperature dependence of $\mathfrak{S}^*(2000)$ is represented by the general form

$$\ln\mathfrak{S}^*(pqrs) = \sum_{i=0}^{n} a_i(\ln T^*)^i, \tag{25}$$

which has proved useful for the correlation of several effective cross sections.[35,42] The coefficients a_i ($i = 0, 1,..., n$) are listed in Table 3. The parameters ϵ/k and σ of the new correlation are identical with those of the earlier fit, but the coefficients are slightly altered. The viscosities generated with this correlation are, therefore, slightly different from those of Ref. 1, but the viscosity deviations are within the accuracy assumed for the latter. The ascribed accuracy for the new correlation is $\pm 0.3\%$ near room temperature, rising to $\pm 0.5\%$ at 1000 K and to $\pm 2\%$ at either extreme of the temperature range (110–2100 K).

The reduced ideal heat capacities C_p^0/R have been taken from the recommendations in the International Union of Pure and Applied Chemistry (IUPAC) tables,[43]

$$C_p^0/R = \sum_{i=1}^{7} f_i T^{i-4} + f_8 u^2 \exp(u)/[\exp(u) - 1]^2, \tag{26}$$

in which

$$u = f_9/T, \tag{27}$$

using the coefficients given in Table 4. The functions (26) and (27), together with the coefficients of Table 4, were optimized[43] to fit quantum-statistical-mechanical calcula-

Table 4.   Coefficients for the representation of the isobaric ideal gas heat capacity $C_p^0(T)$

f_i	Nitrogen[43] Eqs. (24)–(27) (120 K $\leq$ T $\leq$ 2100 K)	Carbon Monoxide Eqs. (35)–(36) (120 K $\leq$ T $\leq$ 2100 K)
f_1	−837.079887373	0.0378038138
f_2	37.91471144874	−0.9247577786
f_3	0.6017378442751	57.301014
f_4	3.504183638234	−255.116608
f_5	−0.8749556530287(−5)	258.012756
f_6	0.1489686072385(−7)	237.03137207
f_7	−0.2563703542771(−11)	−119.9761047
f_8	1.00773735767	3.5
f_9	3353.4061	−17.8

(−n) means 10^{-n}

tions of the heat capacity using spectroscopic data, the results of which were tabulated by Hilsenrath et al.[50] These tabulations are the basis of all modern representations of the ideal-gas heat capacity of nitrogen.

So far as the collision number for internal energy relaxation ζ_{int} is concerned its temperature dependence can be obtained from the thermomolecular pressure difference measurements performed by Millat et al.[42] The internal contribution to the isochoric heat capacity c_{int} is composed of vibrational (vib) and rotational (rot) contributions which have different collision numbers associated with them, ζ_{vib} and ζ_{rot}. But for nitrogen

$$c_{vib}/\zeta_{vib} \ll c_{rot}/\zeta_{rot}, \tag{28}$$

so that if we make use of the approximate result that[42]

$$c_{int}/\zeta_{int} = c_{rot}/\zeta_{rot} + c_{vib}/\zeta_{vib}, \tag{29}$$

we can write

$$\zeta_{int} = \frac{c_{int}}{c_{rot}} \zeta_{rot}. \tag{30}$$

Thus we can derive ζ_{int} solely from a knowledge of the contributions of various modes to the heat capacity and the rotational energy collision number ζ_{rot}. The latter has been shown[42] to be well represented by the equation proposed by Brau and Jonkman[44]

$$\zeta_{rot} = \zeta_{rot}^{\infty}/f(T^*), \tag{31}$$

with

$$f(T^*) = 1 + \frac{\pi^{3/2}/2}{T^{*1/2}} + \frac{2 + \pi^2/4}{T^*} + \frac{\pi^{3/2}}{T^{*3/2}}, \tag{32}$$

where ζ_{rot}^{∞} is a disposable parameter determined by fitting to the thermomolecular pressure data,[42] to be $\zeta_{rot}^{\infty} = 13.384$. For our purposes it is more convenient to employ, in place of ζ_{int}, the corresponding effective cross section $\mathfrak{S}(0001)$ which can then be written

$$\mathfrak{S}(0001) = 0.638\ 28(MT)^{1/2}(c_{rot}/c_{int})f(T^*)/\eta. \tag{33}$$

Here M is the molecular mass and $\mathfrak{S}(0001)$ is given in 10^{-20} m².

Finally, we employ for the term $(\Delta\lambda_{\parallel}/\lambda)_{sat}$, the value determined by Hermans et al.[24]

$$(\Delta\lambda_{\parallel}/\lambda)_{sat} = -0.007\ 77. \tag{34}$$

This value is assumed to be temperature independent in accord with experimental observations.[45]

4.2. Carbon monoxide

That the viscosity of carbon monoxide is essentially identical with that of nitrogen has been confirmed by experiment[46,47] and a recent corresponding states analysis[48] (It is necessary to note here that we make use of the correlations given in Ref. 48 rather than the tabulations given in the article. This is because discussions with the authors of that paper have revealed a systematic error in the tabulations amounting to 0.33%.) Therefore, we have adopted Eqs. (21)–(25) and the coefficients for $\mathfrak{S}^*(2000)$ given in Table 3 for the representation of the viscosity of carbon monoxide.

Goodwin[49] proposed an equation for the isobaric ideal-

gas heat capacity of carbon monoxide in the temperature range 68–1000 K, which was optimized to represent the tabulations of Hilsenrath et al.[50] of statistical-mechanical calculations of C_p^0. The same tabulations have been used to extend Goodwin's function up to 2100 K. Our result is:

$$C_p^0/R = f_8 + \exp(f_9/X)\left(\sum_{i=1}^{7} f_i X^{2-i}\right), \tag{35}$$

with

$$X = T/100.0. \tag{36}$$

The coefficients f_i $(i = 1,...9)$ are summarized in Table 4.

For the representation of ζ_{int} we have employed exactly the same methodology as for nitrogen. Thus, using the thermomolecular pressure difference data of Millat et al.[42] we find for carbon monoxide $\zeta_{rot}^{\infty} = 11.208$ so that

$$\mathfrak{S}(0001) = 0.379\ 17(MT)^{1/2}\frac{c_{rot}}{c_{int}}\frac{f(T^*)}{\eta}, \tag{37}$$

with $f(T^*)$ given by Eq. (32).

For the fractional change in the thermal conductivity of carbon monoxide induced by a magnetic field, we employ the value determined by Hermans et al.[24]

$$(\Delta\lambda_{\parallel}/\lambda)_{sat} = -0.008\ 09. \tag{38}$$

4.3. Results of the analysis

4.3.a. Nitrogen

The theoretical analysis of Sec. 3, together with the sources of experimental data identified earlier, enable the evaluation of a complete set of consistent cross sections $\mathfrak{S}(pqrs)$ as well as D_{int}/D for both nitrogen and carbon monoxide. Figure 1 shows the derived values of D_{int}/D for nitrogen using the various primary data sources for the thermal conductivity. The sensitivity of D_{int}/D to the thermal conductivity data is apparent from the large scatter in this diagram, but there is a sufficiently clear trend to be able to assert that D_{int}/D does indeed approach unity from below in accord with theoretical expectation. This confirms, ex post, the
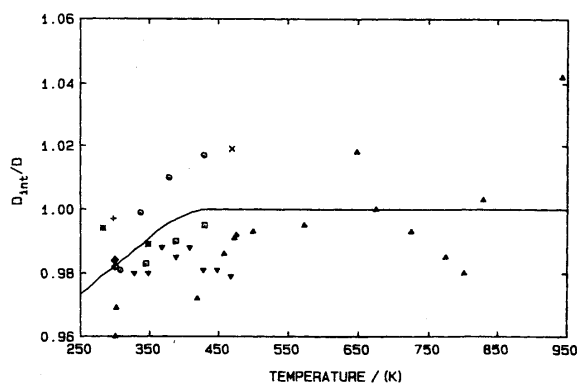

FIG. 1. The ratio of the diffusion coefficient for internal energy to that for mass, D_{int}/D, for nitrogen (O, Ref. 9; □ Ref. 10; × Ref. 11; ◊ Ref. 13; ▼ Ref. 14; ♦ Ref. 16; * Ref. 17; + Ref. 18; ▲ Ref. 19).
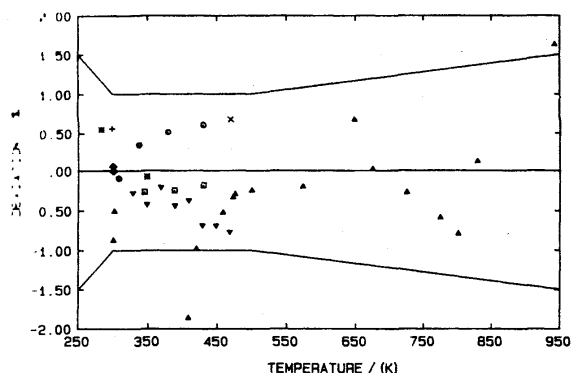
FIG. 2.Deviation plot for the primary thermal conductivity data set for nitrogen (O, Ref. 9; □ Ref. 10; × Ref. 11; ◇ Ref. 13; ▼ Ref. 14; ◆ Ref. 16; * Ref. 17; + Ref. 18; ▲ Ref. 19).

FIG. 3.Deviation plot for the selected secondary thermal conductivity data for nitrogen (+ Ref. 20; × Ref. 21; * Ref. 22; ▲ Ref. 23; ▼ Ref. 14; ◆ Ref. 26; ■ Ref. 27; — Ref. 3; - - - - Ref. 25; - - Ref. 28; · · · · Ref. 30; - - - - Ref.29).

selection of the primary thermal conductivity data since it is consistent with the theoretical results.

Each of the derived cross sections $\mathfrak{S}(0001)$ and $\mathfrak{S}(1001)$ as well as the ratio D_{int}/D have been represented by a correlation of the form of Eqs. (22) and (25) using the reduction parameters of Eq. (24). The coefficients of the optimum correlation are collected in Table 3, together with those for $\mathfrak{S}(2000)$. In the case of D_{int}/D the representation has been chosen so that $D_{int}/D = 1$ for $T \geqslant 430$ K as shown by the solid line in Fig. 1.

The representations of the effective cross sections contained in Eqs. (22) to (25) and Table 3, when combined with a representation of the ideal-gas heat capacity, are sufficient to permit an evaluation of the thermal conductivity of nitrogen over a wide range of temperature. Indeed, this could form the final recommended correlation, except for the fact that it is somewhat cumbersome. Figure 2 compares the results of the calculation of the thermal conductivity by this route with the primary experimental data. It can be seen that the maximum deviation amounts to only ± 1.9% for the data of Le Neindre[19] and that generally the deviations are commensurate with the uncertainty ascribed to the primary data. The solid lines in Fig. 2 delineate the estimated error in the calculated thermal conductivity.

Figure 3 contains a comparison of the calculated thermal conductivity with the results selected from among the secondary data set and with some earlier correlations. As has already been shown for carbon dioxide,[34,51] the data of Saxena and his co-workers[21,22] show a markedly different temperature dependence of the thermal conductivity from that characteristic of the primary data set and that required by theory. Most of the earlier empirical and semiempirical correlations[27-30] show a trend similar to that of Saxena's data because they placed a heavy reliance on that source. The present correlation based upon new, accurate measurements in conjunction with theory must enjoy a higher level of confidence.

4.3.b. Carbon monoxide

Only five sets of thermal conductivity data have been classified as primary data for this gas and the temperature
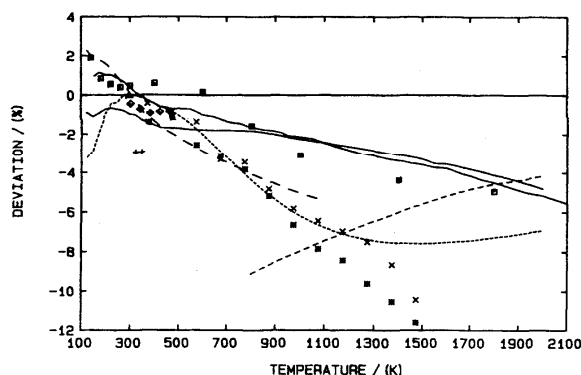
range is limited to $120 \leqslant T \leqslant 430$ K. We have repeated exactly the process described above for nitrogen and evaluated the various effective cross sections $\mathfrak{S}(pqrs)$ and the ratio D_{int}/D. Figure 4 contains a plot of the latter quantity as a function of temperature which shows that, again, the ratio approaches unity from below as the temperature is increased reaching that value at ~ 485 K. Again D_{int}/D has been represented by means of an equation of the form of Eq. (25) for $T \leqslant 485$ K and $D_{int}/D = 1$ for $T > 485$ K. The coefficients in the selected representation of D_{int}/D are contained in Table 3, together with those for the representation of the reduced, effective cross sections $\mathfrak{S}^*(0001)$ and $\mathfrak{S}^*(1001)$. Again these representations, together with that for the heat capacity, enable the thermal conductivity of carbon monoxide to be evaluated over a wider temperature range than is covered by direct, primary measurements. Figure 5 contains a plot of the deviations of the primary experimental thermal conductivity from the calculated values. Above 300 K there is very good agreement with the accurate transient hot-wire data. Below 300 K the experimental data of Johnston and Grilly[32] depart from the correlation by as much as 2%. However, this is not in-

FIG. 4.The ratio of the diffusion coefficient for internal energy to that for mass, D_{int}/D, for carbon monoxide (O Ref. 8; ● Ref. 31; ◇ Ref. 13; * Ref. 16; ▲ Ref. 32).
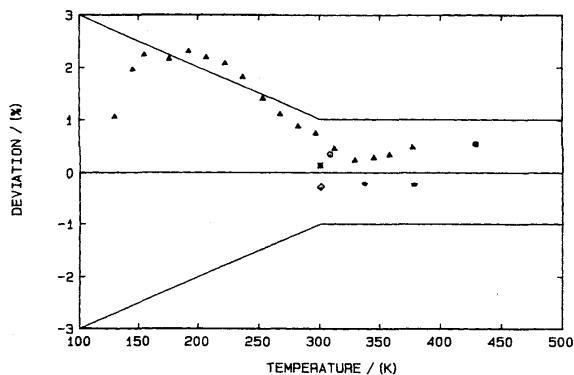
FIG. 5.Deviation plot for the primary thermal conductivity data for carbon monoxide (O Ref. 8; ● Ref. 31; ◊ Ref. 13; * Ref. 16; ▲ Ref. 32).

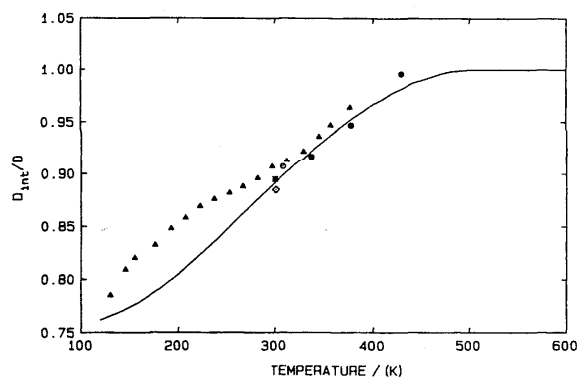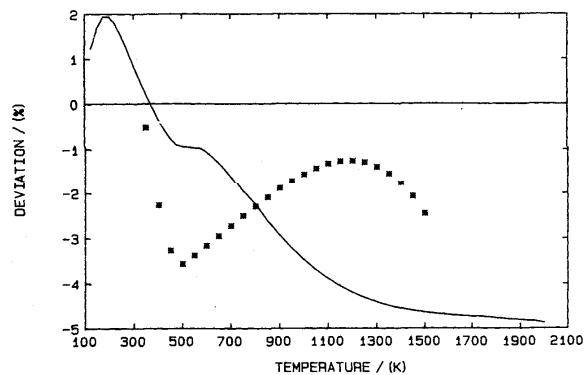FIG. 6.Deviation plot for the selected secondary thermal conductivity data for carbon monoxide (* Ref. 33; — Ref. 29).

consistent with a realistic assessment of the error in these experimental data. For this reason, the solid lines in Fig. 5 which delineate the estimated error in the calculated thermal conductivity rise to $\pm 3\%$ at the lowest temperatures.

Figure 6 compares the secondary experimental thermal conductivity data[33] with the calculated values and includes a comparison with the recent correlation of Matsunaga et al.[29] There are evidently significant differences between the earlier correlation and the high-temperature data, and between the present calculation and the two sets of earlier results.

Owing to the fact that the present calculations are founded on a combination of accurate experimental data and sound theory, they are to be preferred. Nevertheless, it is clear that there is considerable scope for improved measurements of the thermal conductivity at the extremes of temperature.

5. The Final Correlation

Although the formalism set out in Secs. 3 and 4 provides a means of calculating the thermal conductivity of ni-

Table 5. Coefficients for the practical correlation* of the thermal conductivity of nitrogen and carbon monoxide in the temperature range (120 K < T < 2100 K) by means of equations (39) and (40)

Coefficient	Nitrogen	Carbon Monoxide
a_0	4.5384086	4.6027178
a_1	−0.71858394	− 0.46386818
a_2	0.74042225	0.096315989
a_3	−0.91728276	−0.27186355
a_4	0.68036729	0.26415878
a_5	−0.27205905	−0.091615525
a_6	0.055323448	0.010736359
a_7	−0.0045342078	0.0

$$* \quad \ell n \left[\mathfrak{S}_T^*(10E) \right] = \sum_{i=0}^{7} a_i \, (\ell n \, T^*)^i$$

... ,·· .ιι .ιιιl ι:ιιlιοιι monoxide from a combination of experi-
·····ιιι .ιl ιl.ιl.ι :ιιιd theory, it does not represent a particularly
·· . ιιι··ιιl lοrιιι for a practical correlation. Consequently,
·· ,ι·· lι ι lιι ;ιdopt as a practical correlation a representation
ι . .,l ·ιιι lhι· approximate, theoretical equation of Thijsse *et*
,ι·ιν··ιι hy lːq. (20), which may be written

$$l \, \backslash\!\backslash).4627(T/M)^{1/2}(1 + r^2)/\pi\sigma^2 \mathfrak{S}_T^*(10E) \qquad (39)$$

ιι ι· ιιιιs οl' ;ι reduced effective cross section

$$\tilde{\uparrow}(10E) = \mathfrak{S}_T(10E)/\pi\sigma^2. \qquad (40)$$

\ ιlιιι·s οl' $\mathfrak{S}_T^*(10E)$ have been deduced from the thermal
·ιιιlιιι·tivity evaluated according to the procedure described
ιιι \ι·ι·s. 3 and 4. Subsequently, the temperature dependence
·ιl $\tilde{\uparrow}(10E)$ has been represented by a correlation of the
lι·ιιιιι οl' Eq. (25), for which the coefficients securing the
·,ιιlιιιιιιιιι fit are collected in Table 5 for both nitrogen and
. ;ιι hοιι monoxide.

Naturally, the calculated data correlated in this way are

not reproduced exactly. However, the maximum deviations
between the correlation and the calculated data are less than
$\pm 0.1\%$ for nitrogen and $< \pm 0.3\%$ for carbon monoxide.
This additional error is a small fraction of the overall uncer-
tainty that we ascribe to the practical correlation of the ther-
mal conductivity which is one of $\pm 1\%$ between 300 and
500 K rising to $\pm 2.5\%$ at either extreme of the temperature
range. It follows that the deviations of the primary experi-
mental thermal conductivity from the practical correlation
are essentially those displayed in Figs. 2 and 5 for nitrogen
and carbon monoxide respectively.

6. Tabulations

The practical correlation described in the previous sec-
tion has been employed to generate a set of recommended
values for the thermal conductivity of nitrogen and carbon
monoxide over the temperature range 120–2100 K. The val-
ues are listed in Table 6 together, with the corresponding

Table 6: The internal heat capacity and thermal conductivi-
======== ty of nitrogen and carbon monoxide.

Temperature (K)	Nitrogen		Carbon monoxide	
	Int. heat capacity (J/K mol)	Therm. cond. (mW/m K)	Int. heat capacity (J/K mol)	Therm. cond. (mW/m K)
220.0	8.32	20.00	8.33	18.67
225.0	8.32	20.40	8.33	19.08
230.0	8.32	20.79	8.33	19.48
235.0	8.32	21.17	8.33	19.89
240.0	8.32	21.56	8.33	20.29
245.0	8.32	21.94	8.33	20.70
250.0	8.33	22.32	8.33	21.10
255.0	8.33	22.70	8.33	21.50
260.0	8.33	23.08	8.34	21.90
265.0	8.33	23.45	8.34	22.29
270.0	8.33	23.82	8.34	22.69
275.0	8.33	24.19	8.34	23.08
280.0	8.33	24.56	8.34	23.47
285.0	8.33	24.92	8.35	23.86
290.0	8.33	25.28	8.35	24.25
295.0	8.34	25.64	8.35	24.63
300.0	8.34	26.00	8.36	25.02
305.0	8.34	26.35	8.36	25.40
310.0	8.34	26.71	8.37	25.78
315.0	8.35	27.06	8.37	26.16
320.0	8.35	27.41	8.38	26.54
325.0	8.35	27.76	8.38	26.91
330.0	8.36	28.10	8.39	27.28
335.0	8.36	28.45	8.40	27.66
340.0	8.37	28.79	8.41	28.03

Table 6:   The internal heat capacity and thermal conductivi-
=======   ty of nitrogen and carbon monoxide -   continued.

| Temperature | Nitrogen | | Carbon monoxide | |
| | Int. heat capacity | Therm. cond. | Int. heat capacity | Therm. cond. |
(K)	(J/K mol)	(mW/m K)	(J/K mol)	(mW/m K)
345.0	8.37	29.13	8.41	28.39
350.0	8.38	29.47	8.42	28.76
355.0	8.39	29.81	8.43	29.13
360.0	8.39	30.14	8.44	29.49
365.0	8.40	30.48	8.46	29.85
370.0	8.41	30.81	8.47	30.21
375.0	8.41	31.14	8.48	30.57
380.0	8.42	31.47	8.49	30.93
385.0	8.43	31.80	8.51	31.28
390.0	8.44	32.13	8.52	31.64
395.0	8.45	32.45	8.54	31.99
400.0	8.46	32.78	8.55	32.34
405.0	8.47	33.10	8.57	32.69
410.0	8.49	33.42	8.59	33.04
415.0	8.50	33.74	8.60	33.39
420.0	8.51	34.07	8.62	33.74
425.0	8.52	34.38	8.64	34.09
430.0	8.54	34.70	8.66	34.43
435.0	8.55	35.02	8.68	34.77
440.0	8.57	35.34	8.70	35.12
445.0	8.58	35.66	8.73	35.46
450.0	8.60	35.97	8.75	35.80
455.0	8.62	36.29	8.77	36.14
460.0	8.63	36.60	8.80	36.48
465.0	8.65	36.92	8.82	36.82
470.0	8.67	37.23	8.84	37.15
475.0	8.69	37.54	8.87	37.49
480.0	8.71	37.85	8.90	37.82
485.0	8.73	38.17	8.92	38.16
490.0	8.75	38.48	8.95	38.49
495.0	8.77	38.79	8.98	38.83
500.0	8.79	39.10	9.01	39.16
510.0	8.84	39.72	9.06	39.82
520.0	8.89	40.34	9.12	40.48
530.0	8.94	40.95	9.18	41.14
540.0	8.99	41.57	9.25	41.79
550.0	9.04	42.18	9.31	42.44
560.0	9.09	42.80	9.38	43.09
570.0	9.15	43.41	9.45	43.74
580.0	9.21	44.03	9.51	44.38
590.0	9.26	44.64	9.58	45.02
600.0	9.32	45.25	9.65	45.66
610.0	9.38	45.86	9.72	46.30
620.0	9.45	46.47	9.80	46.94
630.0	9.51	47.08	9.87	47.57

Table 6: The internal heat capacity and thermal conductivi-
======= ty of nitrogen and carbon monoxide - continued.

Temperature (K)	Nitrogen		Carbon monoxide	
	Int.heat capacity (J/K mol)	Therm.cond. (mW/m K)	Int.heat capacity (J/K mol)	Therm.cond. (mW/m K)
640.0	9.57	47.69	9.94	48.20
650.0	9.64	48.30	10.01	48.83
660.0	9.70	48.91	10.09	49.46
670.0	9.77	49.52	10.16	50.09
680.0	9.84	50.13	10.23	50.71
690.0	9.90	50.73	10.31	51.33
700.0	9.97	51.34	10.38	51.95
710.0	10.04	51.95	10.46	52.57
720.0	10.10	52.55	10.53	53.19
730.0	10.17	53.16	10.60	53.80
740.0	10.24	53.76	10.68	54.42
750.0	10.31	54.37	10.75	55.03
760.0	10.38	54.97	10.82	55.64
770.0	10.45	55.57	10.90	56.25
780.0	10.51	56.17	10.97	56.85
790.0	10.58	56.78	11.04	57.46
800.0	10.65	57.38	11.11	58.06
810.0	10.72	57.97	11.18	58.66
820.0	10.78	58.57	11.25	59.26
830.0	10.85	59.17	11.32	59.86
840.0	10.92	59.77	11.39	60.46
850.0	10.98	60.36	11.46	61.05
860.0	11.05	60.96	11.53	61.65
870.0	11.11	61.55	11.59	62.24
880.0	11.18	62.14	11.66	62.83
890.0	11.24	62.73	11.72	63.42
900.0	11.31	63.32	11.79	64.01
910.0	11.37	63.91	11.85	64.59
920.0	11.43	64.50	11.92	65.18
930.0	11.49	65.09	11.98	65.76
940.0	11.56	65.67	12.04	66.34
950.0	11.62	66.25	12.10	66.92
960.0	11.68	66.84	12.16	67.50
970.0	11.74	67.42	12.22	68.08
980.0	11.80	68.00	12.28	68.66
990.0	11.85	68.58	12.34	69.23
1000.0	11.91	69.16	12.40	69.81
1010.0	11.97	69.73	12.46	70.38
1020.0	12.03	70.31	12.51	70.95
1030.0	12.08	70.88	12.57	71.52
1040.0	12.14	71.45	12.62	72.09
1050.0	12.19	72.03	12.67	72.66
1060.0	12.25	72.59	12.73	73.23
1070.0	12.30	73.16	12.78	73.79
1080.0	12.35	73.73	12.83	74.36

Table 6:   The internal heat capacity and thermal conductivi-
========   ty of nitrogen and carbon monoxide - continued.

Temperature (K)	Nitrogen		Carbon monoxide	
	Int.heat capacity (J/K mol)	Therm.cond. (mW/m K)	Int.heat capacity (J/K mol)	Therm.cond. (mW/m K)
1090.0	12.40	74.30	12.88	74.92
1100.0	12.46	74.86	12.93	75.48
1110.0	12.51	75.42	12.98	76.05
1120.0	12.56	75.99	13.03	76.61
1130.0	12.61	76.55	13.08	77.16
1140.0	12.66	77.10	13.12	77.72
1150.0	12.71	77.66	13.17	78.28
1160.0	12.75	78.22	13.22	78.83
1170.0	12.80	78.77	13.26	79.39
1180.0	12.85	79.33	13.31	79.94
1190.0	12.89	79.88	13.35	80.50
1200.0	12.94	80.43	13.39	81.05
1210.0	12.98	80.98	13.44	81.60
1220.0	13.03	81.53	13.48	82.15
1230.0	13.07	82.07	13.52	82.70
1240.0	13.11	82.62	13.56	83.25
1250.0	13.16	83.16	13.60	83.79
1260.0	13.20	83.71	13.64	84.34
1270.0	13.24	84.25	13.68	84.88
1280.0	13.28	84.79	13.72	85.43
1290.0	13.32	85.33	13.75	85.97
1300.0	13.36	85.87	13.79	86.51
1310.0	13.40	86.40	13.83	87.05
1320.0	13.44	86.94	13.86	87.59
1330.0	13.48	87.47	13.90	88.13
1340.0	13.52	88.01	13.93	88.67
1350.0	13.55	88.54	13.97	89.21
1360.0	13.59	89.07	14.00	89.75
1370.0	13.63	89.60	14.04	90.28
1380.0	13.66	90.13	14.07	90.82
1390.0	13.70	90.65	14.10	91.35
1400.0	13.73	91.18	14.13	91.88
1410.0	13.77	91.71	14.17	92.42
1420.0	13.80	92.23	14.20	92.95
1430.0	13.83	92.75	14.23	93.48
1440.0	13.87	93.27	14.26	94.01
1450.0	13.90	93.79	14.29	94.54
1460.0	13.93	94.31	14.32	95.06
1470.0	13.96	94.83	14.35	95.59
1480.0	14.00	95.35	14.37	96.11
1490.0	14.03	95.87	14.40	96.64
1500.0	14.06	96.38	14.43	97.16
1510.0	14.09	96.90	14.46	97.69
1520.0	14.12	97.41	14.48	98.21
1530.0	14.15	97.92	14.51	98.73

Table 6: The internal heat capacity and thermal conductivi-
======== ty of nitrogen and carbon monoxide - continued.

Temperature (K)	Nitrogen		Carbon monoxide	
	Int.heat capacity (J/K mol)	Therm.cond. (mW/m K)	Int.heat capacity (J/K mol)	Therm.cond. (mW/m K)
1540.0	14.18	98.43	14.54	99.25
1550.0	14.21	98.94	14.56	99.77
1560.0	14.23	99.45	14.59	100.28
1570.0	14.26	99.96	14.61	100.80
1580.0	14.29	100.47	14.64	101.32
1590.0	14.32	100.98	14.66	101.83
1600.0	14.34	101.48	14.69	102.35
1610.0	14.37	101.99	14.71	102.86
1620.0	14.40	102.49	14.73	103.37
1630.0	14.42	103.00	14.76	103.88
1640.0	14.45	103.50	14.78	104.39
1650.0	14.47	104.00	14.80	104.90
1660.0	14.50	104.51	14.83	105.40
1670.0	14.52	105.01	14.85	105.91
1680.0	14.55	105.51	14.87	106.42
1690.0	14.57	106.01	14.89	106.92
1700.0	14.59	106.50	14.91	107.42
1710.0	14.62	107.00	14.93	107.92
1720.0	14.64	107.50	14.95	108.42
1730.0	14.66	108.00	14.97	108.92
1740.0	14.69	108.49	15.00	109.42
1750.0	14.71	108.99	15.02	109.92
1760.0	14.73	109.48	15.04	110.41
1770.0	14.75	109.98	15.06	110.90
1780.0	14.77	110.47	15.07	111.40
1790.0	14.80	110.97	15.09	111.89
1800.0	14.82	111.46	15.11	112.38
1810.0	14.84	111.95	15.13	112.87
1820.0	14.86	112.44	15.15	113.35
1830.0	14.88	112.93	15.17	113.84
1840.0	14.90	113.43	15.19	114.33
1850.0	14.92	113.92	15.21	114.81
1860.0	14.94	114.41	15.22	115.29
1870.0	14.96	114.90	15.24	115.77
1880.0	14.98	115.39	15.26	116.25
1890.0	15.00	115.88	15.28	116.73
1900.0	15.01	116.36	15.29	117.20
1910.0	15.03	116.85	15.31	117.68
1920.0	15.05	117.34	15.33	118.15
1930.0	15.07	117.83	15.34	118.62
1940.0	15.09	118.32	15.36	119.09
1950.0	15.10	118.81	15.38	119.56
1960.0	15.12	119.29	15.39	120.02
1970.0	15.14	119.78	15.41	120.49
1980.0	15.16	120.27	15.43	120.95

Table 6:   The internal heat capacity and thermal conductivi-
========   ty of nitrogen and carbon monoxide - continued.

| Temperature | Nitrogen | | Carbon monoxide | |
| | Int. heat capacity | Therm. cond. | Int. heat capacity | Therm. cond. |
(K)	(J/K mol)	(mW/m K)	(J/K mol)	(mW/m K)
1990.0	15.17	120.76	15.44	121.41
2000.0	15.19	121.24	15.46	121.87
2010.0	15.21	121.73	15.47	122.33
2020.0	15.22	122.22	15.49	122.79
2030.0	15.24	122.70	15.51	123.24
2040.0	15.25	123.19	15.52	123.69
2050.0	15.27	123.68	15.54	124.14
2060.0	15.29	124.16	15.55	124.59
2070.0	15.30	124.65	15.57	125.04
2080.0	15.32	125.14	15.58	125.48
2090.0	15.33	125.62	15.60	125.93
2100.0	15.35	126.11	15.61	126.37

values of the internal contribution to the isochoric heat capacity. The tabulated values are subject to the uncertainties discussed in Sec. 5 and should be read in conjunction with the error limits displayed in Figs. 2 and 5.

It is interesting to note that the thermal conductivity of carbon monoxide is predicted to differ substantially from that of nitrogen, particularly in the low-temperature region. This is shown in Fig. 7 where the deviation of the thermal conductivity of carbon monoxide from that of nitrogen is plotted. At a temperature of 220 K the difference amounts to almost 6%. This is in contrast to the behaviour of the viscosity of the two gases which are essentially indistinguishable.[47,48]
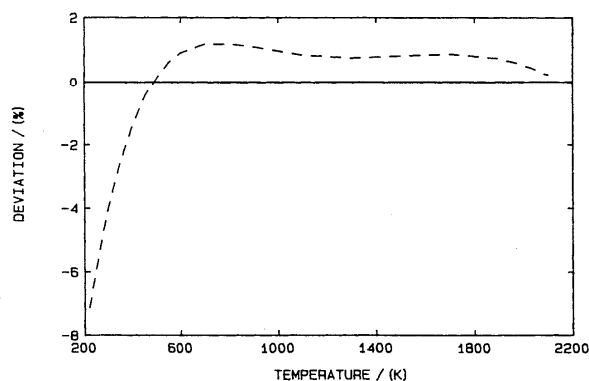

FIG.7.Differences between the thermal conductivities of nitrogen and carbon monoxide (— nitrogen; - - - - carbon monoxide).

7. Conclusions

A concise representation of the thermal conductivity of nitrogen and carbon monoxide in the limit of zero density, based upon a limited set of accurate experimental data and the kinetic theory, has been presented. For either gas the accuracy of the correlation of the thermal conductivity and the associated tabulations is estimated as one of ± 1% in the temperature range 300–500 K, rising to ± 2.5% at either extreme of the temperature range.

8. Acknowledgments

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