Thermal Conductivity of Diatomic Gases: Liquid and Gaseous States

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After an extensive literature survey the experimental thermal-conductivity data for twelve diatomic gases were utilized to produce an accurate and expedient means of predicting values over extensive ranges of temperature and pressure. Plotting values of k^* against T_R on logarithmic coordinates produced similarities pointing to the existence of corresponding states behavior for this family of substances with the exception of hydrogen. Because hydrogen cannot be included in a correlation generalized for the diatomic gases, it has been eliminated from this study. Based on atmospheric pressure data, ratios of $k^*/k^*_{T_c}$ produced a unique relationship with reduced temperature. To include the effect of pressure, residual thermal conductivities were correlated with density for nitrogen and oxygen, the only substances for which high-pressure data exist. These relationships enabled the determination of the thermal conductivity at the critical point. When the value k_c = 8.55 × 10^{-5} cal./sec. cm. °K. for nitrogen was used, an extensive reduced thermal-conductivity correlation was constructed against reduced temperature for parameters of constant reduced pressure. This chart, extending to reduced pressures of 100 and to reduced temperatures of 85, is recommended for the diatomic gases in their gaseous and liquid states.

The developed correlation reproduces experimental nitrogen data to within 1.39%. For the other diatomic gases experimental agreement extends from 1.00 to 3.20%. Such agreement indicates that this correlation is more reliable for the diatomic gases than are other generalized plots presented in the literature.

Present technological advances continue to depend upon progress in understanding the fundamentals of mass and energy transfer; consequently, more careful scrutiny of these operations has stimulated an increasing interest in transport properties. An accurate but simple means of predicting coefficients of diffusion, thermal conductivity, and viscosity is needed to meet the demands of current engineering practice. Besides proving useful in practical applications, a reliable method of determination will be of value in testing and extending modern theories associated with transport phenomena.

Although the use of experimental data is most desirable, such direct values are limited to a few substances. Furthermore, even for the more common substances difficulties inherent in obtaining experimental thermal conductivities restrict the majority of values to the low-pressure region. Where experimental data are lacking, theoretical and empirical equations have been presented (*12, 23, 31, 56*). For accurate usage these are limited in application to the conditions of temperature and pressure for which they have been developed; furthermore, the nature of these expressions makes them inconvenient for expedient usage. Hence, graphical correlations would be desirable, but certain of these (*9*) are too limited to be useful over wide ranges of temperature and pressure. Other methods (*19*) are too generalized to be accurate for specific substances. To overcome these limitations, thermal-conductivity and viscosity correlations have been developed from experimental data specific to families of substances (*7, 42, 48, 51*). Such treatment permits the prediction of accurate values over wide ranges of temperature and pressure for both the gaseous and liquid states. As a continuation of these studies, the available experimental data for the thermal con-

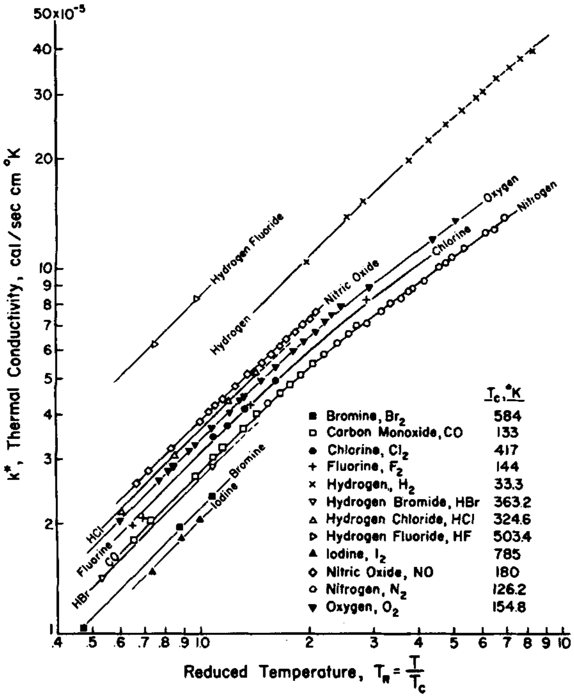

Fig. 1 Thermal conductivity-temperature relationships of diatomic gases at atmospheric pressure.

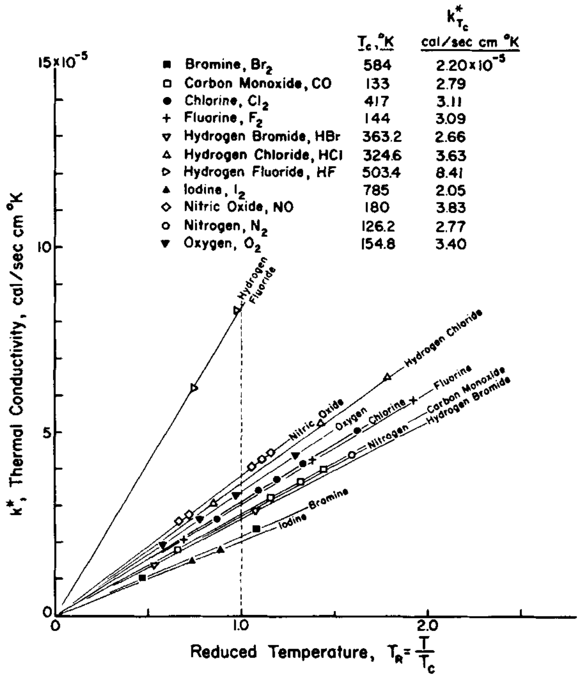

Fig. 2. Thermal conductivity-temperature relationships of diatomic gases at atmospheric pressure.

TABLE 1. COMPARISON OF EXPERIMENTAL THERMAL CONDUCTIVITIES WITH VALUES OBTAINED FROM REDUCED STATE CORRELATION AND RANGE INVESTIGATED

		Deviation, %		Range investigated		
	z_c	This investigation	Owens and Thodos (*42*)	T_R	P_R	References
Bromine	0.3065	1.31	—	0.47-1.88	0.0008-0.0013	*15,17*
Carbon monoxide	0.2943	1.04	2.4	0.59-2.83	0.0003-0.0290	*2,6,11,14,20,21,28,29*
Chlorine	0.2758	2.76	5.6	0.48-1.62	0.0009-0.0131	*14,15*
Fluorine	—	3.12	—	0.65-6.94	0.0017-0.0060	*15,17*
Hydrogen bromide	—	1.98	—	0.26-1.60	0.0020-0.0039	*15*
Hydrogen chloride	0.2656	1.13	3.4	0.61-1.78	0.0034-0.0042	*15*
Hydrogen fluoride	—	1.15	—	0.53-1.56	0.0007-0.0070	*16,18*
Iodine	—	1.93	—	0.57-0.88	0.0006-0.0020	*15*
Nitric oxide	0.2513	1.00	1.00	0.67-2.09	0.0156	*2,14,27,28*
Nitrogen	0.2915	1.39	2.6	0.51-8.50	0.0001-74.1	*2,4,5,6,11,13,14,15, 20,21,22,24,26,30, 32,33,34,36,37,39, 41,43,45,46,49,53, 54,55,58,59,60,61, 63,64*
Oxygen	0.2935	1.67	2.7	0.43-5.08	0.0003-1.93	*2,4,5,11,13,14,18, 20,21,22,27,28,29, 33,40,44,53,58,62, 63,65*

ductivity of diatomic gases have been compiled for the purpose of developing a reduced-state correlation (*47*). This correlation is generalized for the diatomic gases and extends over temperatures ranging from $T_R = 0.3$ to $T_R = 85$ and pressures extending up to $P_R = 100$.

PREVIOUS CORRELATING PROCEDURES

Early attempts at predicting transport properties were based on kinetic theory considerations. Maxwell (*38*) showed the consequences of treating molecules as point centers of forces which vary inversely as the fifth power of the distance between them. Chapman (*8*), assuming only spherical molecules, derived a thermal-conductivity expression which was limited by the accuracy of values for heat capacity and viscosity. Eucken (*13, 14*) proposed a relationship to account for the effect of internal energy in polyatomic molecules.

Sutherland (*57*) proposed an equation for the thermal conductivity at atmospheric pressure as an explicit function of temperature, and Enskog (*12*) and Keyes (*31*) developed the Sutherland type of relationships based on data in limited temperature intervals.

Hirschfelder, Curtiss, and Bird (*23*) present a first approximation to the thermal conductivity of monatomic gases:

$$[k]_1 = 19.89 \times 10^{-5} \frac{\sqrt{T/M}}{\sigma^2 \Omega^{(2,2)*}[T_N]} \quad (1)$$

For the inert gases Owens and Thodos (*42*) found good agreement between experimental values and the results obtained with Equation (1). For polyatomic molecules Equation (1) must be corrected for the transfer of energy between translational and internal degrees of freedom. The correction factor to be applied to Equation (1) is $1 + \{2/5(\rho D/\mu[(2/3)(C_v/R) - 1]\}$. For the restricted condition of $\rho D/\mu = 1$ this factor reduces to the Eucken correction, and the modified form of Equation (1) for polyatomic gases becomes

$$[k_1]^{\text{Eucken}} = \left[\frac{4}{15}\frac{C_v}{R} + \frac{3}{5}\right][k_1] \quad (2)$$

An attempt to predict thermal conductivity at higher pressures follows from the theory of Enskog (*12*). For a gas composed of molecules that are mutually attracting rigid spheres, Enskog presents a power-series function in density to relate the thermal conductivity at high pressures to that at low pressures. A simpler power-series type of expansion has been used by Michels and Botzen (*39*) to account for pressure effects at isothermal conditions.

The correlating methods discussed possess certain serious limitations. The application of their equations must be confined to the temperature range for which they have been developed, and the majority of these expressions are restricted to moderate pressures. Furthermore these equations are not expedient to use, since the refinements which have increased their accuracy limit their ease of application.

Comings and Nathan (*9*) present a simple graphical correlation for the prediction of thermal conductivity for gases at high pressures, which was developed by the use of viscosity and PVT-data for nitrogen, argon, and methane in the Enskog equation. Calculated thermal-conductivity ratios k/k^* were plotted against reduced pressure with parameters of constant reduced temperature. Although later modified and extended by Lenoir, Junk, and Comings (*37*), the final correlation is limited to temperatures between $T_R = 1.0$ and 3.0 and pressures of $P_R < 7.0$.

A more extensive correlation was developed by Gamson (*19*). This represents an initial attempt toward generalization by means of the concept of k_R. This correlation extends to temperatures of $T_R = 10$ and pressures of $P_R = 30$; however no consideration is given to the liquid state. Owing to an overgeneralized approach embodying data for twenty-seven gases, the correlation is claimed to be reliable to within only 15%.

Owens and Thodos (*42*) have developed a reduced-state correlation for the liquid and gaseous states based solely on experimental data for the inert gases. By restricting their method of approach to a family of similar substances, they presented a final correlation that was capable of reproducing the experimental data to within 2%. In view of these improved results, an analogous approach has been adopted for the investigation of the diatomic gases.

CORRELATION OF DATA

An extensive literature survey was undertaken to compile the available thermal-conductivity data of the diatomic gases. The ranges of temperature and pressure and sources of experimental data are presented in Table 1.

Based only upon the atmospheric data, Figure 1 presents a log-log plot of experimental values of k^* as a function of reduced temperature $T_R = T/T_c$. This figure also indicates the twelve gases considered in this study. The notable characteristic of this plot is the parallelism exhibited by the relationships of the various gases. An exception to this is noted for hydrogen at higher temperatures. As postulated from kinetic theory, the behavior of corresponding states should apply to a family of substances possessing molecules of similar shapes and having the same intermolecular-force relationships. The parallelism noted suggests that the behavior of corresponding states should apply to diatomic gases with the exception of hydrogen. An explanation for this anomalous behavior has been advanced in terms of quantum deviations by de Boer and Bird (*10*). For the hydrogen data to conform with those of the other diatomic gases, an adjustment of the critical constants has been utilized by previous investigators. Owing to their empirical nature, these adjustments vary between

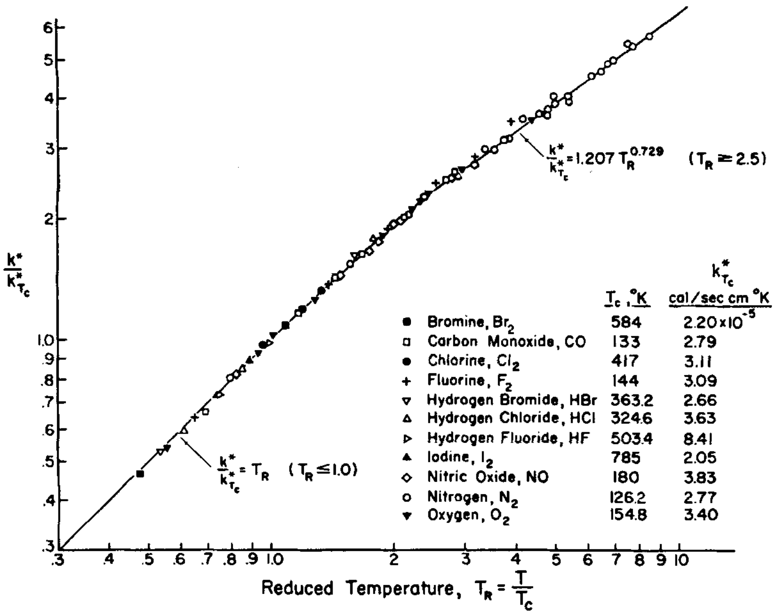

Fig. 3. Ratio of thermal conductivity-temperature relationship for diatomic gases at atmospheric pressure.

investigators and are applicable in limited ranges of temperature and pressure. A more direct and reliable approach is to consider the properties of hydrogen separately from those of the diatomic gases (*48*); consequently, hydrogen has been eliminated from further consideration in these studies.

The similarity of the relationships shown in Figure 1 suggests the representation of these curves by an equation of the form

$$\log k^* = n \log T_R + \log H \qquad (3)$$

For the boundary condition of $T_R = 1$, k^* equals $k_{T_c}{}^*$. Thus Equation (3) becomes

$$\log \frac{k^*}{k_{T_c}{}^*} = n \log T_R \qquad (4)$$

Equation (4) indicates that a unique relationship exists for the diatomic gases when the ratio $k^*/k_{T_c}{}^*$ is related to T_R. This requires a knowledge of $k_{T_c}{}^*$ values specific to each substance.

To obtain $k_{T_c}{}^*$ values more accurately than could be obtained from Figure 1, advantage is taken of the linearity that exists at low temperatures, when k^* is related to temperature on rectilinear coordinates. Therefore the low-temperature thermal-conductivity data for these gases are plotted in Figure 2 as a function of T_R. The values of $k_{T_c}{}^*$ along with the critical temperatures are presented in Figure 2.

An attempt to correlate $k_{T_c}{}^*$ values as a function of molecular weight and the critical values T_c, P_c, and z_c has been made. These studies indicate that $k_{T_c}{}^*$ is independent of z_c. Trial-and-error procedures using the critical constants resulted in the expression

$$k_{T_c}{}^* = 3.05 \times 10^{-5} \frac{P_c{}^{9/16}}{M^{\frac{1}{2}} T_c{}^{1/6}} \qquad (5)$$

Equation (5) is specific to diatomic gases and reliable to 3.4%.

The $k_{T_c}{}^*$ values from Figure 2 and the experimental k^* values were used to calculate ratios of $k^*/k_{T_c}{}^*$, which were plotted against reduced temperature to produce the curve of Figure 3. This unique relationship for the diatomic gases is defined by Equation (4). The relationship is linear below $T_R = 1.0$ and above $T_R = 2.5$ and can be expressed as

$$\frac{k^*}{k_{T_c}{}^*} = T_R \qquad (T_R \leq 1.0) \qquad (6)$$

$$\frac{k^*}{k_{T_c}{}^*} = 1.207 T_R{}^{0.729} \qquad (T_R \geq 2.5) \qquad (7)$$

The correlations presented in Figures 1, 2, and 3 can be used to account for the variation in thermal conductivity with temperature at atmospheric pressure. Sherratt and Griffiths (*50*) state that thermal conductivity is independent of pressure from 1 atm. down to 5 cm. of mercury. Spencer-Gregory and Marshall (*52*, *53*) indicate that the lower limit is between 0.49 and 1.1 cm. of mercury. For pressures above 1 atm. thermal conductivity is dependent not only on temperature but also on pressure.

While extensive data are available at 1 atm., experimental values at higher pressures are limited owing to inherent difficulties in obtaining such values. Of the eleven gases considered in this study nitrogen and oxygen are the only ones for which sufficient data exist to permit the development of a correlation over extensive conditions of temperature and pressure. Even for nitrogen and oxygen too many high-pressure data are lacking to permit this development by direct cross-plotting techniques.

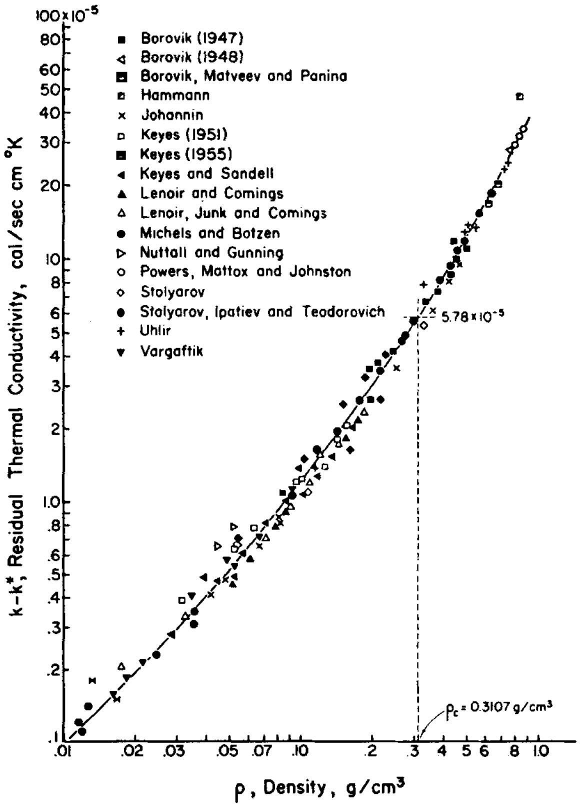

Fig. 4. Residual thermal-conductivity–density relationship for nitrogen in the liquid and gaseous states.

To handle the high-pressure data presently available, use was made of an extension to the method of Abas-Zade (*1*). His correlations, developed from the consideration of functions of invariant quantities, relate thermal conductivity to density and suggest correlating the residual thermal conductivity $k - k^*$ with the density to cover the liquid and gaseous states. Thermal conductivities at atmospheric pressure were substracted from high-pressure thermal conductivities at the same temperature and plotted against density to produce the relationship presented in Figure 4 for nitrogen and Figure 5 for oxygen. The advantage of these correlations is twofold. First is the ability to bring together as a continuous curve the high-pressure gaseous data (*25, 26, 39*) and the low-temperature liquid data (*43, 59*); secondly, the thermal conductivity at any condition of temperature and pressure can be obtained from a knowledge of ρ and k^*. In particular k_c can be evaluated.

When one uses in Figure 4 a value of $\rho_c = 0.3107$ g./cc. (*35*) for the critical density of nitrogen, the residual thermal conductivity at the critical point becomes $k_c - k_{T_c}^* = 5.78 \times 10^{-5}$ cal./sec. cm. °K. Adding to this value $k_{T_c}^* = 2.77 \times 10^{-5}$ cal./sec. cm. °K., one can determine the critical thermal conductivity to be 8.55×10^{-5} cal./sec. cm. °K. Likewise when one uses in Figure 5 a value of $\rho_c = 0.430$ g./cc. (*51*) for the critical density of oxygen, the residual thermal conductivity at the critical point becomes $k_c - k_{T_c}^* = 7.00 \times 10^{-5}$ cal./sec. cm. °K. Adding to this value $k_{T_c}^* = 3.40 \times 10^{-5}$ cal./sec. cm. °K., one can determine the critical thermal conductivity to be 10.40×10^{-5} cal./sec. cm. °K. These critical values permit the construction of reduced thermal-conductivity correlations.

The liquid and high-pressure-gaseous experimental data are more plentiful for nitrogen than for oxygen; moreover, the nitrogen data are considered more reliable. Therefore preference has been given to this substance for the development of a reduced-state correlation applicable to the diatomic gases.

The division of atmospheric thermal conductivities by k_c enabled the construction of the base isobar, $P_R \approx 0$. For pressures above 1 atm. densities at specific conditions of temperature and pressure were used to obtain residual thermal conductivities $k - k^*$ from Figure 4. Values of k^* at the specified temperatures were added to these residuals to produce thermal conductivities corresponding to the density conditions of temperature and pressure. Dividing these values of k by k_c, one obtained reduced thermal conductivities for pressures above 1 atm. These reduced values of k_R were plotted against reduced temperature for parameters of constant reduced pressure. The final correlation is presented in Figure 6. It extends from the high-temperature gaseous region, through the critical point, and into the liquid state. This correlation, based only on experimental nitrogen data, enables the determination of reduced thermal conductivities for diatomic gases from atmospheric pressure to $P_R = 100$ and for temperatures up to $T_R = 85$.

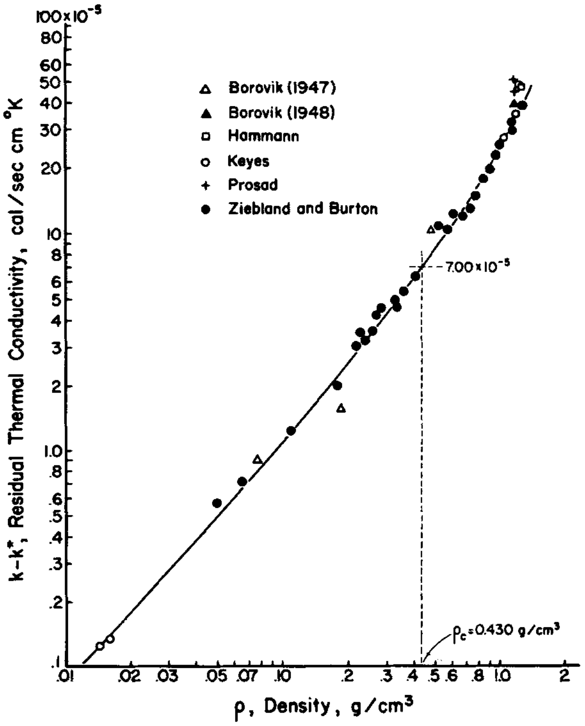

Fig. 5. Residual thermal-conductivity–density relationship for oxygen in the liquid and gaseous states.

To obtain absolute thermal conductivities requires a critical value for each substance. Since experimental data are limited to atmospheric pressure for the diatomic gases other than nitrogen and oxygen, correlations similar to Figures 4 and 5 cannot be developed. Hence k_c cannot be established from experimental data in a manner similar to that used for nitrogen and oxygen. Instead critical thermal conductivities can be obtained by the division of k_R values from Figure 6 into experimental values which are available only at atmospheric pressure.

A more rigorous approach is based on the concept of corresponding states as postulated by the parallel curves of Figure 1. The analytical expression for these relationships is given by Equation (3). Substracting $\log k_c$ from both sides yields

$$\log \frac{k^*}{k_c} = n \log T_R + \log \frac{H}{k_c} \qquad (8)$$

or

$$\log \frac{k^*}{k_c} = n \log T_R + \log \frac{1}{A} \qquad (9)$$

As before, at $T_R = 1.0$, $k^* = k_{T_c}^*$, and therefore

$$\frac{k_{T_c}^*}{k_c} = \frac{1}{A} \qquad (10)$$

or

$$k_c = A k_{T_c}^* \qquad (11)$$

With the use of the values $k_c = 8.55 \times 10^{-5}$ cal./sec. cm. °K. and $k_{T_c}^* = 2.77 \times 10^{-5}$ cal./sec. cm. °K., the ratio $k_c/k_{T_c}^* = 3.087$ was calculated for nitrogen. Likewise with the use of the values $k_c = 10.4 \times 10^{-5}$ cal./sec. cm. °K. and $k_{T_c}^* = 3.40 \times 10^{-5}$ cal./sec. cm. °K., the ratio $k_c/k_{T_c}^* = 3.059$ was calculated for oxygen. The close agreement of these two results validates the use of Equation (11) to calculate values of k_c for the other diatomic substances. Since Figure 6 has been developed from nitrogen data, the value of $A = 3.087$ was selected; it is in contrast to the value of 2.60 proposed by Gamson (*19*) for the twenty-seven gases included in that study. From this value in Equation (11) and the values of $k_{T_c}^*$ obtained from Figure 2, critical thermal conductivities were calculated for the other diatomic gases. These values are presented in Figure 6 along with the critical temperatures and pressures.

DISCUSSION OF RESULTS

The reduced-state correlation presented in Figure 6 has been recommended for the prediction of thermal conductivities for the diatomic gases in the gaseous and liquid states. Its utility depends upon the consistency with which it reproduces experimental data. To be useful, it must be reliable in determining thermal conductivities not only for nitrogen, for which it was developed, but also for the other diatomic gases for which it is recommended. Accordingly thermal conductivities at the experimental conditions of temperature and pressure have been calculated by means of this correlation for purposes of comparison with the experimental data.

Consideration of one hundred fifty nitrogen values representing the available ranges of temperature and pressure produced agreement to within 1.39%. The causes for the average deviation lie in the experimental measurements as well as in the techniques utilized to construct the final correlation. Inherent difficulties are encountered at high pressures. This is evidenced by the lack of accurate data over extensive ranges of temperature and pressure. Even at atmospheric pressure and 273°K. the usually reliable data of Keyes (*30, 32, 34*) are found to disagree within themselves by as much as 2.95%.

In an attempt to produce a reliable correlation, consideration has been given to all the available nitrogen data. Other than the elimination of those investigators whose data were obviously out of line from the general trends, no discrimination has been made. Such elimination is justified by the fact that deviations of 30% were found for the data of Hammann (*22*) and 7.9% for those of Borovik *et al.* (*4, 5, 6*).

To test the usefulness of Figure 6 as a correlation generalized for the diatomic gases, comparisons with available experimental data have been made in a manner similar to that employed for nitrogen. Agreement to within 1.67% was obtained for oxygen in the gaseous state. While this deviation is not large, it does not include the liquid data of Hammann (*22*) and Ziebland and Burton (*65*). Their results deviate by 6.03 and 16.95% respectively. Disagreement of this magnitude was also encountered in viscosity studies (*7*) and, as pointed out by Bird (*3*), can be explained as the result of paramagnetic effects associated with liquid oxygen.

The results of comparing values calculted from Figure 6 with experimental data for the remaining diatomic gases considered in this study are summarized in Table 1, which also presents the deviations for nitrogen and oxygen and those obtained by use of the inert gas correlation of Owens and Thodos (*62*). It is to be noted that except for nitrogen and oxygen these deviations are for the gaseous data available only at atmospheric pressure. The extent of liquid data is limited to the values reported by Borovik, Matveev, and Panina (*6*) for carbon monoxide; their results were found to agree to within 3.30%. From the results of the comparisons presented for the eleven gases considered in this study, it can be concluded that Figure 6 not only reproduces the experimental nitrogen data from which it was developed but is also reliable as a generalized correlation for the other diatomic gases. This is valid provided these substances do not possess anomalous behavior as encountered for liquid oxygen.

As indicated early in this study, hydrogen deviated from a corresponding-states behavior; hence it cannot be included in the generalized reduced state correlation of Figure 6. Consideration has been given to z_c as a possible parameter for correlating $k_c/k_{T_c}^*$ ratios. Because high-pressure thermal-conductivity data are available for hydrogen (*48*), values of k_c and $k_{T_c}^*$ were obtained in a manner analogous to that used for nitrogen and oxygen. These values as well as z_c and the product $k_c/k_{T_c}^*\ z_c$ for these three gases are as follows:

	$k_{T_c}^*$	k_c	$k_c/k_{T_c}^*$	z_c	$k_c/k_{T_c}^*\ z_c$
Hydrogen	5.40×10^{-5}	15.9×10^{-5}	2.944	0.3045	0.8964
Nitrogen	2.77	8.55	3.087	0.2915	0.8999
Oxygen	3.40	10.4	3.059	0.2935	0.8978

If these substances adhere strictly to the behavior of corresponding states, values for the ratio $k_c/k_{T_c}^*$ should be identical, as postulated from Equation (11), but they are not. Multiplying these ratios by their respective critical compressibility factors, one can produce values which are essentially the same. From the average of these values, the following relationship is proposed:

$$\frac{k_c}{k_{T_c}^*} z_c = 0.8980 \qquad (12)$$

This equation is recommended to account for deviations from the behavior of corresponding states for the diatomic gases.

Since values of k_c for the other diatomic gases have been calculated from Equation (11) with $A = 3.087$ resulting from $k_c/k_{T_c}^*$ for nitrogen, it would be meaningless to use these values in Equation (12). Hence until more direct means become available for obtaining k_c values from experimental data the validity of Equation (12) cannot be tested rigorously.

ACKNOWLEDGMENT

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NOTATION

A = constant, k_c/H
C_v = heat capacity at constant volume
H = constant
k = thermal conductivity, cal./sec. cm. °K.
k^* = thermal conductivity at moderate pressures (1 atm.), cal./sec. cm. °K.
k_c = critical thermal conductivity, cal./sec. cm. °K.
k_R = reduced thermal conductivity, k/k_c
$k_{T_c}^*$ = thermal conductivity at the critical temperature and atmospheric pressure, cal./sec. cm. °K.
M = molecular weight
n = exponent, $\varphi(T_R)$
P = pressure, atm.
P_c = critical pressure, atm.
P_R = reduced pressure, P/P_c
R = gas constant
T = absolute temperature, °K.
T_c = critical temperature, °K.
T_N = normalized temperature, $T/(\epsilon/\kappa)$
T_R = reduced temperature, T/T_c
z_c = critical compressibility factor

Greek Letters

ϵ = maximum energy of attraction of two molecules for the Lennard-Jones potential function, erg.
κ = Boltzmann constant, 1.3805×10^{-16} ergs/°K.
ρ = density, g./cc.
ρ_c = critical density, g./cc.
σ = collision diameter for the Lennard-Jones potential function, Å.
$\Omega^{(2,2)*}[T_N]$ = collision integral for Lennard-Jones potential function

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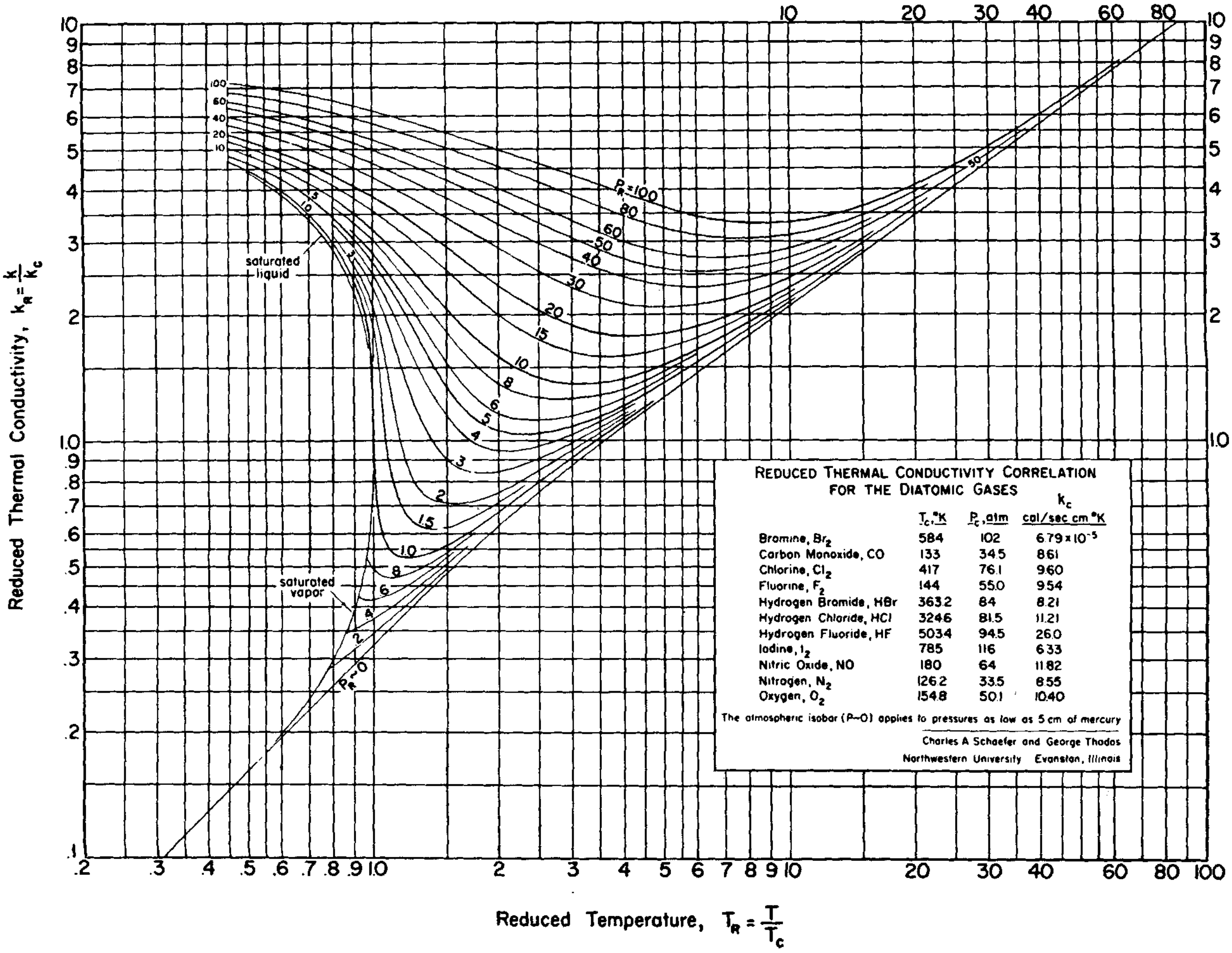

REDUCED THERMAL CONDUCTIVITY CORRELATION FOR THE DIATOMIC GASES

	T_c, °K	P_c, atm	k_c cal/sec cm °K
Bromine, Br_2	584	102	6.79×10^{-5}
Carbon Monoxide, CO	133	34.5	8.61
Chlorine, Cl_2	417	76.1	9.60
Fluorine, F_2	144	55.0	9.54
Hydrogen Bromide, HBr	363.2	84	8.21
Hydrogen Chloride, HCl	324.6	81.5	11.21
Hydrogen Fluoride, HF	503.4	94.5	26.0
Iodine, I_2	785	116	6.33
Nitric Oxide, NO	180	64	11.82
Nitrogen, N_2	126.2	33.5	8.55
Oxygen, O_2	154.8	50.1	10.40

The atmospheric isobar (P~0) applies to pressures as low as 5 cm of mercury

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Fig. 6. Reduced thermal-conductivity correlation for the diatomic gases.

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