

Reprint No. 1 from

Journal of
**Physical and
Chemical
Reference Data**

1972
Volume 1, No. 1, pages 3-118

Gaseous Diffusion Coefficients

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Published by the **American Chemical Society**
and the **American Institute of Physics** for
the **National Bureau of Standards**

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Diffusion coefficients of binary mixtures of dilute gases are comprehensively compiled, critically evaluated, and correlated by new semi-empirical expressions. There are seventy-four systems for which the data are sufficiently extensive, consistent and accurate to allow diffusion coefficients to be recommended with confidence. Deviation plots are given for most of these systems. Almost every gaseous diffusion coefficient which was experimentally determined and reported prior to 1970 can be obtained from the annotated bibliography and table of gas pairs.

A detailed analysis of experimental methods is given, and intercomparison of their results helps establish reliability limits for the data, which depend strongly on temperature. Direct measurements are supplemented by calculations based on knowledge of intermolecular forces derived from independent sources—molecular beam scattering for high temperatures, and London dispersion constants for low temperatures. In addition, diffusion coefficients for several mixtures are obtained from experimental data on mixture viscosities and thermal diffusion factors. Combination of all these results gives diffusion coefficients over a very extensive temperature range, from very low temperatures to 10 000 K.

All data are corrected for composition dependence and for quantum effects. New semi-empirical equations are derived for making such corrections easily.

Key words: Binary gas mixtures; critically evaluated data; diffusion; diffusion coefficients; gases; transport properties.

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1. Introduction

The purpose of this report is to evaluate published data on gaseous diffusion coefficients and establish recommended values when possible. Short catalogs of measured diffusion coefficients have previously appeared in the International Critical Tables [1],¹ the Landolt-Börnstein Tables [2], and the Thermophysical Properties Research Literature Retrieval Guide [3]. Additional limited reviews are also available [4-8]. In the present survey gaseous diffusion coefficients are comprehensively compiled and critically evaluated, including analysis for consistency with theory and with other measured molecular properties. Such detailed evaluation is possible because of the existence of a highly developed theory for dilute gases.

The scope of this survey is therefore limited to the dilute gas region. In this region the density is such that the rates of transport of mass, momentum, and energy are entirely controlled by binary molecular collisions. In practice this means gases with densities approximately corresponding to

standard conditions (i.e., of the order of $10^{19} - 10^{20}$ molecules per cm³). The emphasis here is on two-component (binary) mixtures. Multicomponent diffusion can be accurately described in terms of the binary diffusion coefficients for all possible pairs of gases in the mixture [9]. All the atoms or molecules considered are neutral species. The mass transport processes outside the scope of this survey are as follows: (1) diffusion of ionized particles (ion mobility), (2) mixture separations in a temperature gradient (thermal diffusion), (3) mixing due to convection or turbulence, and (4) the permeation of gases through liquids or solids. The sole interest is the mixing of gases caused by composition gradients.

The gaseous diffusion coefficients recommended here are succinctly reported by means of semi-empirical functions; temperature limits range up to 10 000 K and to a lower temperature of the order of 100 K (tables 12 and 13). There is a small composition dependence in the diffusion coefficients which may be estimated from parameters listed in table 15. Figures 5 to 81 are deviation plots, for sixty-two gas pairs, of experimental measurements from the semi-empirical equations; these graphs illustrate discrepancies in the data.

The procedure used in this report is as follows. At all possible temperatures published diffusion coefficient values were critically evaluated on an individual gas-pair basis. The data assessment was determined without any additional experimental measurements. From the rigorous kinetic theory of gases an approximation was developed to make corrections for small composition effects. Coefficients could then be normalized to a specific mixture concentration for comparison and subsequent correlation. Diffusion coefficients derived from other transport property measurements, particularly mixture viscosities, were useful for the extension of values to intermediate temperatures and for consistency checks. In the absence of direct measurements, intermolecular forces from theory and from beam experiments served to determine diffusion coefficients at very low and elevated temperatures, respectively. Semi-empirical functions were constructed to correlate the data over three decades of temperature within the experimental uncertainty.

This report is divided into five major sections. It begins with a section—Theoretical Background—which includes the diffusion coefficient definition and its theoretical expression according to the rigorous kinetic theory of gases. The kinetic-theory foundations are necessary for the understanding of temperature and composition dependences, and quantum effects. This section closes with equations for the determination of diffusion coefficients from intermolecular forces and from other transport properties. The principal experimental techniques are described next. Methods of measurement are classified by the geometry of the apparatus, and their reliability is estimated. Procedures used to critically evaluate the entire body of experimental data for accuracy, composition and temperature dependencies are outlined under Treatment of Data. The analyses and results related to the small composition dependence of the diffusion coefficient are entirely in this section. The semi-empirical correlation equation was chosen on the basis of knowledge

¹ Figures in brackets indicate the literature references at the end of Section 1.

of how intermolecular forces affect the temperature dependence of the diffusion coefficients. Previously uncalculated values of low-temperature asymptotes of diffusion coefficients are tabulated.

The fourth major section—Results—can be subdivided into four areas. First, diffusion-coefficient uncertainty limits are classified according to temperature and gas pair. Second, the tabulation of correlation parameters for the recommended data is given. Then a series of graphs shows the relative deviations between the recommended coefficients and the data. An inspection of these graphs will readily indicate that the unqualified selection of a diffusion coefficient from the literature may be uncertain by at least several percent. The last part of the Results section contains detailed remarks about data appraisals for specific systems. The final major part of this report is the Bibliography; two annotated bibliographies are given; one contains all the experimental sources, complete through 1968, according to author (gas pair and method are noted), and the second is a supplementary listing of citations according to gas pair. Practically all diffusion coefficients ever measured can be traced through these bibliographies; however, for many systems the results are too fragmentary or too uncertain for the diffusion coefficients

to be accepted as reliable. Additional references from 1969, 1970, and a few from 1971 are included, but the correlation is complete only through 1968.

References for Section 1

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- [9] Hirschfelder, J. O., Curtiss, C. F., and Bird, R. B., *Molecular Theory of Gases and Liquids* (John Wiley and Sons, New York, 1964), pp. 487, 517.

2. Theoretical Background

In this section the presentation of the theoretical background is preceded by the phenomenological definition of diffusion coefficients. Then in outline form expressions for diffusion coefficients are derived by the Chapman-Enskog procedure for a solution of the Boltzmann equation. Most mathematical details of the derivation are omitted, and the discussion accentuates the application limits of these rigorous kinetic-theory formulas. More complete information is available in three monographs [1–3],² and recent developments appear in several articles [4–13]. The emphasis here is on molecular physics as a prerequisite to the understanding of accepted theoretical results which are necessary in order to achieve the following:

- (1) suggest the mathematical form for the correlation of diffusion coefficients as a function of temperature,
- (2) correct diffusion coefficient measurements for composition dependence,
- (3) estimate quantum effects for low-temperature diffusion coefficients,
- (4) calculate diffusion coefficients directly from knowledge of intermolecular forces, and
- (5) calculate diffusion coefficients from other transport properties.

2.1. Phenomenological Definition of the Gaseous Diffusion Coefficient

In a nonuniform mixture the diffusion coefficient is a proportionality constant between the molecular flux and the composition gradient of a species. Diffusion coefficients are defined by phenomenological equations for two-component and multi-component mixtures.

a. Two-Component Mixtures

In two-component mixtures, in the absence of temperature and pressure gradients, external forces, and chemical reactions, the flux equations are

$$\mathbf{J}_1 = -n\mathcal{D}_{12}\nabla x_1, \quad (2.1-1)$$

$$\mathbf{J}_2 = -n\mathcal{D}_{21}\nabla x_2. \quad (2.1-2)$$

Each species (or component) is labeled by subscripts 1 or 2. The flux densities are \mathbf{J}_1 and \mathbf{J}_2 (molecules/cm²·s), the total number density is n (molecules/cm³), and the composition gradients are in terms of mole fractions x_1 and x_2 . These equations hold only in the case of zero net flux, $\mathbf{J}_1 + \mathbf{J}_2 = 0$. If the net flux is not zero, eqs (2.1-1) and (2.1-2) can be considered to hold in a coordinate system moving with the net flux, that is, at a velocity equal to $(\mathbf{J}_1 + \mathbf{J}_2)/n$.

The diffusion coefficients \mathcal{D}_{12} and \mathcal{D}_{21} are positive constants with units of cm²/s. It is easy to show from eqs (2.1-1) and (2.1-2) that $\mathcal{D}_{12} = \mathcal{D}_{21}$, because $\mathbf{J}_1 + \mathbf{J}_2 = 0$ and $x_1 + x_2 = 1$ for a binary mixture. Thus diffusion in a binary mixture is described by a single diffusion coefficient.

Molecular diffusion, strictly speaking, cannot occur under conditions in which both the net flux and the pressure gradient are simultaneously zero. If the pressure is uniform, then in general fluxes are different for different species, and the net flux is not zero. If the net flux is zero, a small pressure gradient must exist in order to counter the tendency for the different species fluxes to be different [14–16]. For instance, in a closed system the difference in the species fluxes causes the number density and hence the pressure to increase on one side of the system and decrease on the other side until the resulting pressure gradient forces the net

² Figures in brackets indicate the literature references at the end of Section 2.

flux to be zero. If the fluxes were to remain unequal in a closed system, then the pressure would continue to increase on one side and decrease on the other side.

The pressure gradients in diffusing gas mixtures turn out to be very small in magnitude, however; in fact, they are almost immeasurably small except in capillary tubes, where they have been measured [17–26]. Because of this, it is unnecessary to include in the flux equations any term directly proportional to a pressure gradient. The whole effect of any pressure gradient is simply to modify the net flux, and this is the only term that needs to be directly included.

The generalization of eqs (2.1–1) and (2.1–2) for nonzero net flux is therefore simply

$$\mathbf{J}_1 = -n\mathcal{D}_{12}\nabla x_1 + x_1\mathbf{J}, \quad (2.1-3)$$

$$\mathbf{J}_2 = -n\mathcal{D}_{21}\nabla x_2 + x_2\mathbf{J}, \quad (2.1-4)$$

where the net flux is $\mathbf{J} = \mathbf{J}_1 + \mathbf{J}_2$. Equations (2.1–3) and (2.1–4) define the diffusion coefficient in a stationary coordinate system. It can be easily shown, as before, that $\mathcal{D}_{12} = \mathcal{D}_{21}$.

The preceding equations, which define binary diffusion coefficients, are applicable to any fluid, and hold regardless of any dependence of the diffusion coefficient on composition, pressure, or temperature. For dilute gases the pressure and composition dependences are especially simple—the diffusion coefficient is inversely proportional to pressure and is only slightly dependent on mixture composition. The temperature dependence is more complicated. All these factors are treated in more detail in subsequent sections.

b. Multicomponent Mixtures

The flux of a species in a multicomponent mixture is not conveniently expressed in terms of composition gradients as in the foregoing equations for binary mixtures. The reason is that the multicomponent diffusion coefficients thereby defined have an excessively complicated composition dependence which makes the calculation of a flux a formidable task. A simpler set of equations for multicomponent diffusion is obtained by a different arrangement of terms—the composition gradient of a species is related to differences in fluxes of gas pairs [2, 3, 8, 27]. The outstanding advantage of such a relationship is a description of multicomponent diffusion in terms of diffusion coefficients for binary mixtures. These equations, credited to Stefan and Maxwell, are

$$\nabla x_i = \sum_{j=1}^{\nu} \frac{n_i n_j}{n^2 \mathcal{D}'_{ij}} \left(\frac{\mathbf{J}_j}{n_j} - \frac{\mathbf{J}_i}{n_i} \right), \quad (2.1-5)$$

where i and j denote the species. For a mixture of ν species there are ν equations, but only $\nu-1$ are independent. The diffusion coefficients \mathcal{D}'_{ij} depend primarily only on the nature of the species i and j , but are not quite the same as the corresponding binary diffusion coefficients \mathcal{D}_{ij} (hence the prime). However, the difference between the \mathcal{D}'_{ij} and the \mathcal{D}_{ij} lies only in their weak composition dependence, the exact value of \mathcal{D}'_{ij} depending slightly on the composition of the whole multicomponent mixture and not on just the relative

amounts of i and j . The variation of the binary \mathcal{D}_{ij} with composition is empirically a few percent at most, and is of the same magnitude as the experimental uncertainties in the few available multicomponent \mathcal{D}'_{ij} , or even in most measured binary \mathcal{D}_{ij} for that matter. Therefore it is reasonable on an empirical basis to take $\mathcal{D}'_{ij} \approx \mathcal{D}_{ij}$ for multicomponent diffusion. This is also justified theoretically, for in the first Chapman-Enskog approximation \mathcal{D}'_{ij} and \mathcal{D}_{ij} are identical and independent of composition [2, 3, 7].

The special case of a trace species diffusing through a uniform multicomponent mixture is of interest for two reasons. It provides a simple test of $\mathcal{D}'_{ij} \approx \mathcal{D}_{ij}$, and makes possible the calculation of diffusion coefficients of various species in air. First, denote the trace species by 1 and assume the absence of a net flux ($\mathbf{J} = 0$), then eqs (2.1–5) reduce to a single equation,

$$\nabla x_1 = -\mathbf{J}_1 \sum_{j=2}^{\nu} \frac{x_j}{n \mathcal{D}'_{1j}}. \quad (2.1-6)$$

If the trace diffusion coefficient \mathcal{D}_1 is defined to be the constant of proportionality between \mathbf{J}_1 and ∇x_1 , then

$$\frac{1}{\mathcal{D}_1} = \sum_{j=2}^{\nu} \frac{x_j}{\mathcal{D}'_{1j}}, \quad (2.1-7)$$

where \mathcal{D}_1 specifies the diffusion coefficient of the tracer in the multicomponent mixture. If \mathcal{D}'_{ij} replaces \mathcal{D}'_{ij} then eq (2.1–7) becomes an expression of Blanc's law [28]. Detailed calculations [29] of \mathcal{D}'_{ij} and \mathcal{D}_{ij} for this special case show that the deviations from Blanc's law are small for ordinary gases. This further justifies the application of binary diffusion coefficients to eq (2.1–5) for multicomponent diffusion. The second case of interest, the calculation of diffusion coefficients of a species in air (when direct measurements are unavailable or inadequate), is especially convenient by the application of Blanc's law with available binary diffusion coefficients of the species in nitrogen and in oxygen.

2.2. Molecular Theory of Diffusion

a. General Background

This presentation of the molecular theory of diffusion briefly outlines some major points of the rigorous kinetic theory of gases. Kinetic theory postulates transport due entirely to molecules in motion. In diffusion the individual molecules themselves carry mass through the gas. Since there are immense numbers of molecules moving about in a gas it is to be expected that molecular encounters (or collisions) are of cardinal importance in controlling the overall rate at which transport occurs. The collisions in turn are controlled by the forces of interaction between the molecules. By the formulas of kinetic theory, knowledge of these fundamental intermolecular forces can lead to gaseous diffusion coefficients.

The importance of molecular collisions in diffusion can be illustrated by some typical numerical values. At ordinary conditions of temperature and pressure molecules in gases have molecular speeds

of the order of 10^4 cm/s, which is about the speed of sound. In contrast, actual diffusion velocities (J_i/n_i) are much less—about 1 cm/s. This great decrease in apparent molecular speed occurs because diffusion is dominated by collisions which cause the molecular paths to be twisted into tortuous shapes. The actual path of a molecule is approximately 10^4 times the net distance traveled during diffusion. For gases at ordinary conditions only binary collisions are important; ternary and higher-order collisions are very unlikely. Binary collisions, two-particle encounters, are characteristic of gases with the ratio of mean free path to molecular diameter of the order of 100.

Transport phenomena—diffusion, viscosity, thermal conductivity, and thermal diffusion—arise by deviations, however slight, from the equilibrium molecular velocity distribution function known as the Maxwell distribution. At equilibrium conditions an isolated gas mixture has no gradients in composition, pressure, or temperature; thus no fluxes. Therefore to obtain transport coefficients on a theoretical basis knowledge of a nonequilibrium velocity distribution function is a necessary requirement.

b. Theoretical Methods

Diffusion coefficients can be calculated from a flux derived from a molecular concept—the integral of molecular velocity over the nonequilibrium velocity distribution function. The velocity distribution function represents the probability for a molecule to have a specific velocity and location at some instant. The changes in the velocity distribution due to molecular interactions must satisfy the nonlinear Boltzmann integrodifferential equation. The basic problem of rigorous kinetic theory is to solve the Boltzmann equation.

A solution of the Boltzmann equation was independently obtained by Chapman and by Enskog [1–3]. Both used a method of successive approximation, and even though procedures by Chapman and Enskog differ in detail the results are identical. The transport properties appear finally in the Chapman-Enskog theory as solutions of infinite sets of simultaneous algebraic equations, and the transport properties can be expressed formally as ratios of infinite determinants whose elements are the coefficients of the algebraic equations. The coefficients of the equations are complicated functions which depend on the species and the composition of the mixture, and on integrals related to binary molecular interactions. These sets of equations can be solved, fortunately, by rapidly converging approximation schemes.

An outline of the Chapman-Enskog procedure is as follows. First the velocity distribution function is expanded in terms of a perturbation function added to the Maxwell (equilibrium) distribution. By the assumption of a small perturbation, the expansion substituted back into the Boltzmann equation leads to a linearized integrodifferential equation for the perturbation (ref. 2, sec. 7.3b, c).

The perturbation term is assumed proportional to gradients, and expanded in a series; the series expansion coefficients are functions of molecular velocities (ref. 2, sec. 7.3d). The assumption of linearity in the gradient of composition is precisely consistent with the preceding phenomenological definition of diffusion coefficients; other transport

coefficients may be accounted for by additional appropriate gradients. The diffusion coefficient now appears as an integral of the expansion coefficient over the molecular velocities (ref. 2, sec. 7.4a). The expansion coefficient satisfies a linear integrodifferential equation obtained from the Boltzmann equation. This equation is solved by a second series expansion in terms of squares of molecular velocities. For the second expansion it is convenient, but not necessary, to use orthogonal functions because orthogonal properties lead to subsequent simplification of the calculations. The orthogonal functions usually used are Sonine polynomials (ref. 2, sec. 7.3d, g). When this second expansion is substituted back into the integral expression for the diffusion coefficient, it turns out (because of the orthogonality) that the diffusion coefficient is exactly equal to just one of the coefficients in the second expansion (ref. 2, sec. 7.4a). The problem now is to find the coefficients of the second expansion. To do this, the expansion is substituted back into the linear integrodifferential equation, which is then solved by a moment or a variational method. The result is an infinite set of algebraic equations in which the unknowns are the coefficients of the second expansion, and the coefficients of these unknowns are complicated multiple integrals over molecular velocities. These integrals result from the moment formation; most of the integrations can be carried out explicitly, but not all, until the law of intermolecular force is specified (ref. 1, chap. 9; ref. 2, sec. 7.4d).

The diffusion coefficient is thus equal to a single unknown in an infinite set of algebraic equations. This set cannot be solved exactly except in very special cases, and some successive approximation procedure must be used. The set is systematically truncated in some plausible way (two ways are commonly used, one due to Chapman and Cowling, and the other to Kihara) [39]; the simplest truncation gives the first approximation to the diffusion coefficient, the next step gives the second approximation, and so on. In the first approximation the diffusion coefficient is independent of composition; the second and higher approximations introduce composition dependence. Since the approximation procedure converges rapidly, the third approximation for the diffusion coefficient is almost identical with the second approximation [30].

The solution of the Boltzmann equation by the Chapman-Enskog procedure depends on the following assumptions:

Binary Collisions. The Boltzmann equation itself has a fundamental assumption—binary collisions. This assumption—that only two-molecule interactions are important—limits the application of theoretical results to transport properties of dilute gases.

Small Mean Free Path. The Chapman-Enskog solution assumes that the dimensions of the gas container are large compared to the molecular mean free path. In gases at extremely low densities molecules collide more frequently with the walls of the container than with each other. When molecular collisions with a container surface are significant, the theory fails.

Small Perturbation. In the Chapman-Enskog theory the assumption of a small perturbation function describes small departures from the equilibrium velocity distribution function; in other words, at

conditions slightly away from equilibrium the transport property fluxes are linear in the gradients.

Classical Mechanics. Historically, classical mechanics was necessarily used by Boltzmann, Chapman, and Enskog; however, their theory can be reformulated to account for quantum-mechanical effects. The modification needed is merely to replace an integration over classical impact parameters for molecular interactions by an integration over deflection angles involving the quantal differential cross section.

Elastic Collisions. The original Boltzmann equation and its solution by Chapman and Enskog were limited to elastic collisions between molecules interacting with central forces. Inelastic collisions occur between molecules with internal degrees of freedom, and kinetic energy is no longer conserved, although mass and momentum are conserved. Thus diffusion and viscosity are not strongly affected by the presence of internal degrees of freedom, but thermal conductivity is. The theory may be reformulated to account for inelastic collisions.

c. Theoretical Results

In this section the Chapman-Enskog theoretical expressions for diffusion coefficients are given, as well as the definition of collision integrals, and a number of complementary definitions related to binary molecular collisions. The extension of the collision integral formulas to include inelastic collision effects is also given.

Approximation Scheme for Diffusion Coefficients. The higher approximations for diffusion coefficients in a dilute gas binary mixture with species of type 1 and 2 are written

$$[\mathcal{D}_{12}]_M = [\mathcal{D}_{12}]_1 f^{(M)}, \quad (2.2-1)$$

where $[\mathcal{D}_{12}]_1$ is the first approximation, $f^{(M)}$ accounts for the effects of higher approximations, and M indicates the order of approximation. In the first approximation for diffusion coefficients, $f^{(1)}=1$; the effect of higher approximations is described by

$$f^{(M)} = 1/(1 - \Delta_{12} - \dots), \quad (2.2-2a)$$

or

$$f^{(M)} = (1 + \Delta_{12} + \dots), \quad (2.2-2b)$$

where Δ_{12} is the first correction term to $[\mathcal{D}_{12}]_1$.

First Approximation for the Diffusion Coefficient. The expression for $[\mathcal{D}_{12}]_1$ is

$$[\mathcal{D}_{12}]_1 = \frac{3}{16} \left(\frac{2\pi kT}{\mu_{12}} \right)^{1/2} \left(\frac{1}{n\bar{\Omega}_{12}^{(1,1)}} \right), \quad (2.2-3)$$

where $\mu_{12} = m_1 m_2 / (m_1 + m_2)$ is the reduced mass of a pair of molecules, m is the molecular mass of a species, k is the Boltzmann constant, and T is the absolute temperature. The diffusion collision integral $\bar{\Omega}_{12}^{(1,1)}$ has units of area and is dependent on the temperature and the forces of molecular interaction of the gas. The collision integral for diffusion is

$$\bar{\Omega}_{12}^{(1,1)}(T) = \frac{1}{2} (kT)^{-3} \int_0^\infty e^{-E/kT} E^2 S^{(1)}(E) dE, \quad (2.2-4)$$

where E is the initial relative translational energy of two molecules in a binary collision, $E = \frac{1}{2} \mu_{12} v^2$, v being the initial relative speed of the molecular

pair, and the diffusion transport cross section is

$$S^{(1)}(E) = 2\pi \int_0^\pi (1 - \cos \chi) I(\chi, E) \sin \chi d\chi, \quad (2.2-5)$$

where $I(\chi, E)$ is the differential scattering cross section. For classical systems $I(\chi, E) \sin \chi d\chi = b db$, where b is the impact parameter—the perpendicular distance between one molecule and the initial line of relative approach of the other molecule. The classical scattering angle for a pair of colliding molecules is

$$\chi = \pi - 2b \int_{r_c}^\infty \frac{dr}{r^2} \left[1 - \left(\frac{b}{r} \right)^2 - \frac{\varphi(r)}{E} \right]^{-1/2}, \quad (2.2-6)$$

where r_c , the distance of closest approach, is given by

$$1 - \left(\frac{b}{r_c} \right)^2 - \frac{\varphi(r_c)}{E} = 0. \quad (2.2-7)$$

In eq (2.2-6) r is the internuclear separation distance, and $\varphi(r)$ is the spherically symmetric intermolecular potential.

The expression for $[\mathcal{D}_{12}]_1$ in practical units is

$$[\mathcal{D}_{12}]_1 = 0.008258 \left(\frac{M_1 + M_2}{2M_1 M_2} \right)^{1/2} \frac{T^{3/2}}{p \bar{\Omega}_{12}^{(1,1)}}, \quad (2.2-8)$$

where T is in degrees Kelvin, p is the pressure in atmospheres, M_1 and M_2 are the molecular weights in grams per mole, and $\bar{\Omega}_{12}^{(1,1)}$ is in angstroms squared.

The first approximation for the diffusion coefficient is independent of mixture composition.

Second Approximation for the Diffusion Coefficient. The second approximation for the diffusion coefficient is

$$[\mathcal{D}_{12}]_2 = [\mathcal{D}_{12}]_1 (1 + \Delta_{12}), \quad (2.2-9)$$

where

$$\Delta_{12} = \frac{(6C_{12}^* - 5)^2}{10} \left(\frac{x_1^2 P_1 + x_2^2 P_2 + x_1 x_2 P_{12}}{x_1^2 Q_1 + x_2^2 Q_2 + x_1 x_2 Q_{12}} \right). \quad (2.2-10)$$

The P 's and Q 's are complicated algebraic expressions which contain various collision integrals and are defined in Section 2.4; C_{12}^* is a collision integral ratio given by eq (2.2-16). The first correction term Δ_{12} is temperature dependent, and contains the small composition dependence of the diffusion coefficient.

Accuracy of Formulas for the Diffusion Coefficient. How close $[\mathcal{D}_{12}]_1$ is to $\lim_{M \rightarrow \infty} [\mathcal{D}_{12}]_M$ depends on

composition, molecular masses, and the intermolecular forces of the gas mixture. Of course, an experiment measures only $\lim_{M \rightarrow \infty} [\mathcal{D}_{12}]_M$. By numerical comparison of $[\mathcal{D}_{12}]_1$, $[\mathcal{D}_{12}]_2$, $[\mathcal{D}_{12}]_3$, etc. for a variety of special cases, the accuracy of $[\mathcal{D}_{12}]_1$ may be assessed [30]. For the case of nearly equal molecular masses $[\mathcal{D}_{12}]_1$ is probably accurate to within 2 percent regardless of the composition or intermolecular forces. If the molecular masses are very unequal and the heavy component is the trace species then $[\mathcal{D}_{12}]_1$ is accurate to within 1 percent.

If the light component is the trace then $[\mathcal{D}_{12}]_1$ may be quite inaccurate; the worst case known is a mixture of rigid spheres for which $[\mathcal{D}_{12}]_1$ is low by about 13 percent. In practical cases it is probably safe to regard $[\mathcal{D}_{12}]_1$ as accurate within about 5 percent for all gas pairs, and $[\mathcal{D}_{12}]_2$ as accurate within 2 percent.

Pressure Dependence of Diffusion Coefficients. All theoretical approximations for dilute-gas diffusion coefficients are inversely proportional to density, or pressure. It can be shown by elementary kinetic theory arguments that the molecular flux is independent of pressure for binary collisions. The reason is that the number of flux carriers (i.e., the molecules) is directly proportional to their number density n , but the number of particles that impede the flux by collisions is also proportional to n . The two effects exactly compensate. If the associated gradient is chosen so as not to involve n , then the constant of proportionality must also be independent of n . Thus the coefficients of viscosity and thermal conductivity are independent of density. But the proportionality constant for diffusion is arbitrarily chosen to be $n\mathcal{D}_{12}$ (for historical reasons), so that \mathcal{D}_{12} itself must be inversely proportional to n .

Collision Integrals for Elastic Collisions. The general equation for collision integrals is

$$\bar{\Omega}^{(l,s)}(T) = [(s+1)!(kT)^{s+2}]^{-1} \int_0^\infty e^{-E/kT} E^{s+1} S^{(l)}(E) dE, \quad (2.2-11)$$

with

$$S^{(l)}(E) = \left[1 - \frac{1 + (-1)^l}{2(1+l)} \right]^{-1} \int_0^{2\pi} d\phi \int_0^\pi (1 - \cos^l \chi) I(\chi, \phi, E) \sin \chi d\chi, \quad (2.2-12)$$

where l and s specify weighting factors related to the mechanism of transport by molecular collisions; χ and ϕ are the polar azimuth angles which describe the orientation of the final relative molecular velocity to the initial relative velocity in a collision. From eq (2.2-3) it is evident that for diffusion $l=1$ and $s=1$; the viscosity and thermal conductivity collision integrals have $l=2$ and $s=2$. Other values of l and s occur only in the expressions for higher approximations. Collision integrals are calculated for realistic intermolecular force models only by difficult numerical integrations (ref. 1, chap. 10; ref. 2, chap. 8).

The definition of collision integrals as dimensionless reduced quantities, that is, collision integrals divided by the analogous quantities for rigid-sphere molecules, makes calculations of transport coefficients more convenient. The reduced collision integral is defined as

$$\Omega^{(l,s)*} \equiv \frac{\bar{\Omega}^{(l,s)}}{\pi\sigma^2}, \quad (2.2-13)$$

where σ is an arbitrary molecular size or range-of-force parameter, and is exactly unity for rigid spheres of diameter σ . Numerical values of reduced collision integrals are usually about unity if σ is chosen in a reasonable way, and differences from unity reflect differences in effective molecular size

for the selected intermolecular force model in comparison to an ideal rigid-sphere model.

In the higher approximations for diffusion coefficients, and in other transport properties as well, several recurring ratios of collision integrals, or reduced collision integrals, are defined for calculation convenience, namely

$$A^* = \Omega^{(2,2)*} / \Omega^{(1,1)*}, \quad (2.2-14)$$

$$B^* = [5\Omega^{(1,2)*} - 4\Omega^{(1,3)*}] / \Omega^{(1,1)*}, \quad (2.2-15)$$

$$C^* = \Omega^{(1,2)*} / \Omega^{(1,1)*}, \quad (2.2-16)$$

$$E^* = \Omega^{(2,3)*} / \Omega^{(2,2)*}. \quad (2.2-17)$$

The magnitude of each of these ratios is approximately unity, and exactly unity for rigid spheres.

Collision integrals and collision integral ratios are functions of temperature and the parameters of the selected model for intermolecular forces. Since such models usually have at least two parameters, one with dimensions of distance and one with dimensions of energy, it is economical to tabulate collision integrals in dimensionless form, in which the reduced collision integral is given as a function of a reduced temperature. Reduced collision integrals have already been defined; reduced temperature is usually defined as

$$T^* \equiv kT/\epsilon, \quad (2.2-18)$$

where ϵ is the energy parameter of the potential (usually the depth of the minimum).

Collision Integrals for Inelastic Collisions. As previously mentioned, the kinetic theory of gas transport properties by the Chapman-Enskog procedure applies strictly to molecules that have no internal degrees of freedom. To extend the preceding equations to polyatomic and polar molecules the theory of transport properties must account for inelastic collisions. This can be achieved only by a reformulation of the Boltzmann equation in which the nonequilibrium velocity distribution function must be specified for all the internal energy states of molecules. A semiclassical treatment is used in which the translational molecular motion is described classically, as before, but the internal motions are described quantum-mechanically. The formal kinetic theory of transport properties that includes inelastic collisions in the Chapman-Enskog scheme was originally developed for pure gases by Wang Chang, Uhlenbeck, and deBoer [31], and by Taxman [32]. Additional theoretical work [7-9, 13] has extended the theory to mixtures; the derived collision integrals correspond to the first approximations of the Chapman-Enskog theory.

The available results for inelastic collision integrals are for the most part formal in the sense that the integrations are too difficult to carry out for realistic models, even with the fastest available computers. But useful conclusions can be drawn from them without going through elaborate calculations. These conclusions are stated at the end of this subsection.

The general equations for the diffusion and viscosity collision integrals are as follows:

$$\bar{\Omega}_{qq'}^{(s)}(T) = 2[(s+1)!Z_q Z_{q'}]^{-1}$$

$$\sum_{ijkl} e^{-\epsilon_{qi} - \epsilon_{q'j}} \int_0^\infty \gamma^{2s+3} e^{-\gamma^2} S_{ij}^{(s)kl}(E) d\gamma, \quad (2.2-19)$$

where

$$\gamma^2 S_{ij}^{(1)kl}(E) = \int_0^{2\pi} d\phi \int_0^\pi I_{ij}^{kl}(\chi, \phi, E) \sin \chi d\chi (\gamma^2 - \gamma\gamma' \cos \chi), \quad (2.2-20)$$

$$\gamma^4 S_{ij}^{(2)kl}(E) = \frac{3}{2} \int_0^{2\pi} d\phi \int_0^\pi I_{ij}^{kl}(\chi, \phi, E) \sin \chi d\chi [\gamma^2 (\gamma^2 - \gamma'^2 \cos^2 \chi) - \frac{1}{6} (\gamma^2 - \gamma'^2)^2], \quad (2.2-21)$$

$$\gamma^2 - \gamma'^2 = \epsilon_{qk} + \epsilon_{q'l} - (\epsilon_{qi} + \epsilon_{q'j}), \quad (2.2-22)$$

$$\gamma^2 = E/kT, \quad (2.2-23a)$$

$$\gamma'^2 = E'/kT, \quad (2.2-23b)$$

in which the prime on γ refers to the relative kinetic energy after a collision and the species are denoted by q and q' . The various ϵ 's are the energies of the internal quantum states of the species, divided by kT . Z_q and $Z_{q'}$ are the internal partition functions for the q and q' species: $Z_q = \sum \exp(-\epsilon_{qi})$ and $Z_{q'} = \sum \exp(-\epsilon_{q'j})$. They appear only as normalization factors in eq (2.2-19). The indices i and j denote the i th and j th internal quantum states of the q th and q' th species before a collision, and k and l the corresponding states after a collision. The differential scattering cross section $I_{ij}^{kl}(\chi, \phi, E)$ describes collisions between two molecules initially in internal states i and j which undergo a collision and finally are in states k and l . In the collision integral of eq (2.2-19) the superscript l is primed so as not to be confused with the l th quantum state.

The collision integrals for inelastic processes reduce exactly to collision integrals for elastic collisions when $E' = E$ and the differential scattering cross section is the same as the elastic cross section, $I_{ij}^{ij} = I_{el}$ for all i and j .

Inelastic collisions enter $\bar{\Omega}^{(1,1)}$ only through the term $\gamma\gamma' \cos \chi$; to a first approximation $\gamma \approx \gamma'$ and the inelastic collisions have no effect. For a second approximation γ' can be written as γ plus some terms in $\Delta\epsilon_{qq'}$, where $\Delta\epsilon_{qq'} = \gamma^2 - \gamma'^2$; the inelastic contributions are then of the form $\gamma(\Delta\epsilon_{qq'}) \cos \chi$. For isotropic molecular scattering the correction term vanishes, and even for nonisotropic scattering the inelastic contribution is probably small unless there is some special correlation between $\Delta\epsilon_{qq'}$ and χ . The $\bar{\Omega}^{(2,2)}$ may also reduce to a manageable form, in the first approximation $\Delta\epsilon_{qq'} \ll \gamma^2$ and the terms in $\Delta\epsilon_{qq'}$ may be dropped. For a second approximation, the terms in $\Delta\epsilon_{qq'}$ vanish for isotropic scattering.

The determination of \mathcal{D}_{12} from mixture viscosity measurements is especially related to A_{12}^* . In these calculations the algebraic expressions (see sec. 2.7) appear mathematically the same whether the molecular collisions are elastic or inelastic. This is important because the only effect depends on what value is substituted for A_{12}^* . A first-order expansion for A_{12}^* indicates only a small correction for inelastic collisions, but good approximations are not yet available.

2.3. Temperature Dependence of Diffusion Coefficients

The temperature dependence of \mathcal{D}_{12} according to the preceding expressions must be investigated in order to develop a general equation useful for the correlation of diffusion coefficients. Almost the entire temperature dependence is given by the factor $[T^{3/2}/\bar{\Omega}^{(1,1)}(T)]$ appearing in $[\mathcal{D}_{12}]_1$; that is, the higher approximations have only a slight effect. Accordingly the temperature dependence of Δ_{12} is disregarded in the following discussion. The temperature dependence of $[\mathcal{D}_{12}]_1$ can be calculated if the law of force between two molecules is known. Details about intermolecular forces will follow later in this section. Calculations for plausible molecular force laws have shown that the derivative $d \ln \bar{\Omega}^{(1,1)}(T)/d \ln T$ usually lies between 0 and $-1/2$, so that the derivative $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$ lies between $3/2$ and 2 . Thus \mathcal{D}_{12} should vary as $T^{3/2}$ to T^2 , and this is usually found to be the case experimentally. These general features are depicted in figure 1.

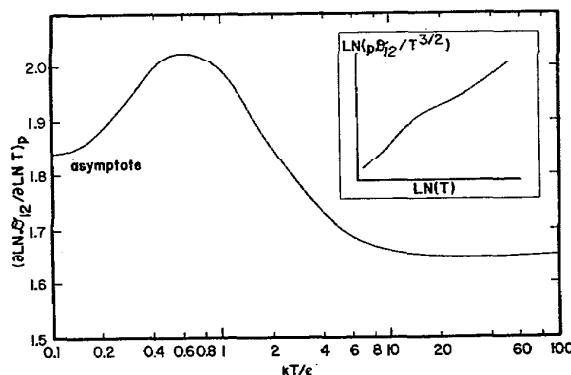


FIGURE 1. Qualitative temperature dependence of diffusion coefficients.

This figure shows the derivative $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$, obtained from experimental data and intermolecular force models. The simple molecular model of ideal rigid spheres, sets a lower bound of $3/2$ to the derivative, independent of temperature. Actual gas pairs, however, have appreciably greater values of the derivative than $3/2$.

The general characteristics of $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$ are as follows. At extremely low temperatures the dominant interaction is the long-range r^{-6} London dispersion energy, which causes $\bar{\Omega}^{(1,1)}(T)$ to vary as $T^{-1/3}$. At extremely high temperatures the dominant interaction is the (roughly) exponential short-range repulsion energy, which causes $\bar{\Omega}^{(1,1)}(T)$ to have a weaker temperature dependence than at low temperatures. Thus $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$ is equal to $11/6$ at low temperatures, and equal to a smaller value, ~ 1.7 , at high temperatures, the high-temperature value being slightly dependent on temperature. In the intermediate temperature region $(\partial \ln \mathcal{D}_{12}/\partial \ln T)_p$ is not monotonic, and exhibits a maximum where both short-range and long-range forces are significant.

In figure 1 the inset shows $\ln(p\mathcal{D}_{12}/T^{3/2})$ versus $\ln T$. This curve illustrates the behavior to be expected from very low temperatures up to about $10\,000$ K, and indicates the form of relationship

$$\bar{\Omega}_{qq'}^{(l',s)}(T) = 2[(s+1)!Z_q Z_{q'}]^{-1} \sum_{ijkl} e^{-\epsilon_{qi} - \epsilon_{q'j}} \int_0^\infty \gamma^{2s+3} e^{-\gamma^2} S_{ij}^{(l')kl}(E) d\gamma, \quad (2.2-19)$$

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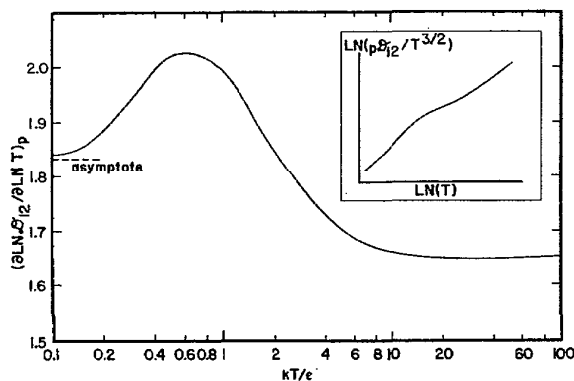


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In figure 1 the inset shows $\ln(p\mathcal{D}_{12}/T^{3/2})$ versus $\ln T$. This curve illustrates the behavior to be expected from very low temperatures up to about 10 000 K, and indicates the form of relationship

needed to fit \mathcal{D}_{12} data as a function of temperature. Possible quantum effects at very low temperatures have been ignored in this illustration since they are important only for a few very light gases.

More quantitative information about the temperature dependence of \mathcal{D}_{12} requires additional details about intermolecular potentials. As is well known, molecules attract each other at large separation distances and repel each other at small separations. In principle, quantum theory provides a method for calculating the interaction between a pair of molecules [33]. The long-range interactions are dominated by London dispersion forces, and can be calculated fairly accurately [34], but the short-range interactions are too complicated to be calculated in any simple way. The various interactions and their effect on \mathcal{D}_{12} are considered below.

a. Long-Range Interactions

These interactions behave asymptotically as (neglecting retardation effects)

$$\varphi(r) = -C/r^6, \quad (2.3-1)$$

where C is the London constant. According to classical mechanics the collision integral has the form

$$\bar{\Omega}^{(1,1)} \propto (C/T)^{1/3}. \quad (2.3-2)$$

Thus as $T \rightarrow 0$, $\mathcal{D}_{12} \propto T^{1/3}$ classically, but at sufficiently low temperatures quantum corrections become important. A general expression for the quantum-mechanical $\bar{\Omega}^{(1,1)}$ as $T \rightarrow 0$ is not presently available.

b. Short-Range Interactions

Short-range interactions can be approximated by an exponential function, and over a more limited range by an inverse power. These single-term potentials have a simple algebraic form which permits the collision integral to be calculated numerically; such results lead to values of \mathcal{D}_{12} at high temperatures, $T \gtrsim 1000$ K.

The expression for the exponential potential is

$$\varphi(r) = \varphi_0 \exp(-r/\rho), \quad (2.3-3)$$

in which φ_0 and ρ are empirical parameters. For this potential the $\bar{\Omega}^{(1,1)}(T)$ has been evaluated [35] over a wide temperature range by numerical methods, and its temperature dependence found to be approximately

$$\bar{\Omega}^{(1,1)}(T) \propto [\ln(\varphi_0/kT)]^2. \quad (2.3-4)$$

Thus at high temperatures diffusion coefficients are expected to be proportional to $T^{3/2}/[\ln(\varphi_0/kT)]^2$.

The inverse-power repulsive potential can be written as

$$\varphi(r) = K/r^s, \quad (2.3-5)$$

where K and s are empirical parameters. For this potential the temperature dependence of the collision integral is exactly [1, 2]

$$\bar{\Omega}^{(1,1)}(T) \propto (sK/kT)^{2/s}. \quad (2.3-6)$$

For this model the diffusion coefficients are proportional to $T^{3/2+2/s}$.

c. Intermediate-Range Interactions

At intermediate internuclear separation distances the potential is not dominated by either attractive or repulsive forces. The potential has a "well" whose detailed shape is not precisely known; descriptive approximations are frequently given by semi-empirical expressions which interpolate between functions derived for solely attractive or repulsive interactions. For spherical nonpolar molecules two such well-known approximations are,

Lennard-Jones ($n-6$)

$$\varphi(r) = \left(\frac{n\epsilon}{n-6}\right) \left[\frac{6}{n} \left(\frac{r_m}{r}\right)^n - \left(\frac{r_m}{r}\right)^6 \right], \quad (2.3-7)$$

and

Exp-6

$$\varphi(r) = \left(\frac{\alpha\epsilon}{\alpha-6}\right) \left\{ \frac{6}{\alpha} \exp\left[\alpha\left(1-\frac{r}{r_m}\right)\right] - \left(\frac{r_m}{r}\right)^6 \right\}, \quad (2.3-8)$$

where ϵ is the depth of the potential energy well, r_m is the location of the potential energy minimum, and n and α are parameters which reflect the steepness of the repulsive forces. Such potentials give a complicated relationship for the temperature dependence of the collision integral, and no analytic expression can be given corresponding to intermediate temperatures (about 200 to 1000 K for most gas pairs). However, Sutherland [36] developed a simple relationship for rigid-sphere molecules with weak attractive interactions, and showed that

$$\Omega^{(1,1)*} = 1 + S/T, \quad (2.3-9)$$

where S is a positive constant. The temperature dependence of \mathcal{D}_{12} is then

$$\mathcal{D}_{12} \propto T^{3/2}/(1+S/T), \quad (2.3-10)$$

which correlates experimental results well over moderate temperature ranges. This form can also accurately represent collision integrals for the Lennard-Jones (12-6) potential (within 0.2% for $1.4 < kT/\epsilon < 3.5$) [37]. Another relationship, suggested by Reinganum [38], is

$$\Omega^{(1,1)*} = e^{S/T}, \quad (2.3-11)$$

or

$$\mathcal{D}_{12} \propto T^{3/2}e^{-S/T}, \quad (2.3-12)$$

which reduces to the Sutherland form for small values of S/T .

2.4. Composition Dependence of Diffusion Coefficients

In this section the theoretical results are given for the small composition dependence of gaseous diffusion coefficients. The composition correction, less than 5 percent for most gas pairs, is needed to eliminate systematic discrepancies in the evaluation and correlation of \mathcal{D}_{12} measurements. The composition correction term, Δ_{12} , is repeated here for con-

venience, and the P and Q terms are expressed as follows:

$$\Delta_{12} = \frac{(6C_{12}^* - 5)^2}{10} \left(\frac{x_1^2 P_1 + x_2^2 P_2 + x_1 x_2 P_{12}}{x_1^2 Q_1 + x_2^2 Q_2 + x_1 x_2 Q_{12}} \right), \quad (2.2-10)$$

where

$$P_1 = \frac{2M_1^2}{M_2(M_1 + M_2)} \left(\frac{2M_2}{M_1 + M_2} \right)^{1/2} \frac{\Omega_{11}^{(2,2)*}}{\Omega_{12}^{(1,1)*}} \left(\frac{\sigma_{11}}{\sigma_{22}} \right)^2, \quad (2.4-1)$$

$$P_{12} = 15 \left(\frac{M_1 - M_2}{M_1 + M_2} \right)^2 + \frac{8M_1 M_2 A_{12}^*}{(M_1 + M_2)^2}, \quad (2.4-2)$$

$$Q_1 = \frac{2}{M_2(M_1 + M_2)} \left(\frac{2M_2}{M_1 + M_2} \right)^{1/2} \frac{\Omega_{11}^{(2,2)*}}{\Omega_{12}^{(1,1)*}} \left(\frac{\sigma_{11}}{\sigma_{12}} \right)^2 \\ \times \left[\left(\frac{5}{2} - \frac{6}{5} B_{12}^* \right) M_1^2 + 3M_2^2 + \frac{8}{5} M_1 M_2 A_{12}^* \right], \quad (2.4-3)$$

$$Q_{12} = 15 \left(\frac{M_1 - M_2}{M_1 + M_2} \right)^2 \left(\frac{5}{2} - \frac{6}{5} B_{12}^* \right) \\ + \frac{4M_1 M_2 A_{12}^*}{(M_1 + M_2)^2} \left(11 - \frac{12}{5} B_{12}^* \right) \\ + \frac{8}{5} \frac{(M_1 + M_2)}{(M_1 M_2)^{1/2}} \frac{\Omega_{11}^{(2,2)*}}{\Omega_{12}^{(1,1)*}} \frac{\Omega_{22}^{(2,2)*}}{\Omega_{12}^{(1,1)*}} \left(\frac{\sigma_{11}}{\sigma_{12}} \right)^2 \left(\frac{\sigma_{22}}{\sigma_{12}} \right)^2. \quad (2.4-4)$$

The relations for P_2 and Q_2 are obtained from those for P_1 and Q_1 by an interchange of subscripts. The subscript "11" denotes molecular interactions between two type 1 molecules, and so on. The Chapman and Cowling relations for the Q 's have been presented, not Kihara's [39].

The above complicated formulas for Δ_{12} are tedious to use, and attempts have been made to simplify the expressions [40-42]. The results are semi-empirical approximations, one of which [42] takes a form that determines the most sensitive parts of Δ_{12} from experiment and the remainder from theoretical calculations. An improved semi-empirical approximation for Δ_{12} is developed in this report; details appear in section 4.2.

2.5. Quantum Effects on Diffusion Coefficients

Quantum effects become significant when the de Broglie wavelength, $\lambda = h/\mu v$, approaches the

size parameter σ . Thus the ratio λ/σ is a measure of quantum effects, and gases behave classically for $\lambda/\sigma \ll 1$. In kinetic theory it is common practice to use the deBoer parameter Λ^* ,

$$\Lambda^* \equiv h/[\sigma(2\mu\epsilon)^{1/2}], \quad (2.5-1)$$

which is simply λ/σ for a colliding pair of reduced mass μ and kinetic energy equal to the depth, ϵ , of the potential well. The larger the value of Λ^* , the more important are the quantum effects at a given reduced temperature, $T^* \equiv kT/\epsilon$. This is illustrated in table 1, which is based on calculations for the Lennard-Jones (12-6) potential [43]. A gas behaves classically at all temperatures for $\Lambda^* = 0$; typical values of Λ^* are as follows: 0.35 for Ne-Ar, 1.3 for He-Ne, 1.5 for H₂-D₂, and 2.9 for ³He-⁴He. From table 1 it is evident that quantum deviations in \mathcal{D}_{12} can be quite large for light gases at low temperatures. However, the collision integral ratio A_{12}^* has deviations of only a few percent, so that reliable values of \mathcal{D}_{12} can be computed from accurate viscosity measurements even when quantum effects are important, as explained in more detail in section 2.7.

The only modification necessary for quantum effects is the replacement of the integration over classical impact parameters by one over the quantum-mechanical differential cross section. The quantum transport cross section as given by a scattering phase-shift analysis is

$$I(\chi) = |f(\chi)|^2, \quad (2.5-2)$$

$$f(\chi) = \frac{1}{2i\kappa} \sum_{l=0}^{\infty} (2l+1) [\exp(2i\delta_l) - 1] P_l(\cos \chi), \quad (2.5-3)$$

in which δ_l is the phase shift, l denotes the angular momentum quantum number, and κ is the wave number of relative motion, equal to $2\pi\mu v/h = 2\pi/\lambda$. The phase shifts are obtained by the solution of the radial wave equation. $P_l(\cos \chi)$ is a Legendre polynomial in $\cos \chi$, and $f(\chi)$ is the scattering amplitude. When eqs (2.5-2) and (2.5-3) are substituted into eq (2.2-12), the integrations can be carried out to yield the following expressions for

TABLE 1. Quantum effects on diffusion coefficients and on collision integral ratio Λ_{12}^* in terms of the deBoer parameter, Λ^* , and the reduced temperature, $T^* \equiv kT/\epsilon^a$

Λ^*		$[\mathcal{D}_{12}]_1$ (Quantal)/ $[\mathcal{D}_{12}]_1$ (Classical)						A_{12}^* (Quantal)/ A_{12}^* (Classical)				
		0	0.5	1.0	1.5	2.0	3.0	0.5	1.0	1.5	2.0	3.0
0.1	1	1.001	1.001	0.641	0.741	2.924	1.014	1.043	0.970	0.921	0.966	
0.2	1	1.004	0.899	.813	1.046	2.494	1.010	1.042	.979	.956	1.046	
0.5	1	1.002	.991	1.069	1.297	1.883	1.007	0.994	.957	.956	1.021	
1.0	1	1.009	1.032	1.105	1.224	1.444	0.999	.984	.971	.978	1.018	
1.5	1	1.008	1.031	1.080	1.150	1.269	.999	.990	.987	.994	1.025	
2.0	1	1.006	1.025	1.060	1.105	1.182	1.000	.994	.996	1.003	1.028	
3.0	1	1.004	1.016	1.035	1.060	1.101	1.001	.997	1.002	1.008	1.027	

^a Calculated from a Lennard-Jones (12-6) potential.

the diffusion and viscosity (or thermal conductivity) transport cross sections:

$$S^{(1)}(E) = \frac{4\pi}{\kappa^2} \sum_{l=0}^{\infty} (l+1) \sin^2(\delta_{l+1} - \delta_l), \quad (2.5-4)$$

and

$$S^{(2)}(E) = \frac{4\pi}{\kappa^2} \sum_l \frac{(l+1)(l+2)}{(2l+3)} \sin^2(\delta_{l+2} - \delta_l). \quad (2.5-5)$$

The summations are over all integral values of l from 0 to ∞ for distinguishable particles, but only over the even or odd integral values for indistinguishable particles (in which case the summation is multiplied by a normalization factor of 2). In order to describe observable processes $S^{(1)}$ must always refer to distinguishable particles, but $S^{(2)}$ can refer to either. These formulas apply only to the case of elastic collisions; corresponding formulas for inelastic collisions have never been derived.

It is often desired to adjust measurements of \mathcal{D}_{12} for a set of isotopes to a common molecular weight basis. This is especially important for hydrogen isotopes (H_2 , D_2 , T_2 , HD, etc.) for which there are many measurements for different isotope pairs. The diffusion coefficient has a mass dependence which may involve three factors. First, the principal dependence of \mathcal{D}_{12} on mass is the proportionality to the inverse square root of the reduced mass of the gas pair. A second mass dependence factor is in the composition correction term Δ_{12} , but this is almost always negligible. The third dependence is in the diffusion collision integral, which in the quantum case depends on mass through the deBoer parameter Λ^* . In order to make the necessary computations a potential model is assumed, and the diffusion collision integral is then obtained for both isotopic mixtures; for the Lennard-Jones (12-6) potential quantum collision integrals have been published [43, 44] as a function of the deBoer parameter and reduced temperature in convenient tabular form. For any two mixtures a simple ratio of $\Omega^{(1,1)*}(\Lambda^*, T^*)$ is taken to adjust the data according to eq (2.2-8). Since this procedure is model-dependent, it is reliable only when the adjustment is small.

It is sometimes useful to express the exact formulas of eqs (2.5-4) and (2.5-5) as semiclassical expansions, in which the leading term is the classical formula and the quantum corrections appear as a series in powers of Planck's constant (or Λ^*). Explicit expressions have been obtained for the first two quantum corrections [45], but little use has yet been made of these results. Most numerical calculations to date have used the exact formulas in terms of phase shifts.

2.6. Determination of Diffusion Coefficients from Intermolecular Forces

In this section expressions for diffusion collision integrals are presented which lead to \mathcal{D}_{12} at conditions unavailable by direct experiment. The expressions for $\bar{\Omega}^{(1,1)}$ are given only for long-range and short-range interactions; for intermediate-range interactions, the $\bar{\Omega}^{(1,1)}$ are not given because the corresponding values of \mathcal{D}_{12} are available by direct experiment. Information about long-

range interactions is obtained from molecular polarizabilities, oscillator strengths, and other optical data; a summary of the various results has been published [34]. The short-range interactions are based on molecular beam scattering experiments [46-48]. For both ranges of interaction the specific data sources used in this report are listed in the Bibliography, sections II and III.

a. Diffusion Collision Integrals for Long-Range Interactions

The collision integral for the London r^{-6} attractive potential is

$$\bar{\Omega}^{(1,1)} = 71.1(C/T)^{1/3}, \quad (2.6-1)$$

in which C is the London constant in atomic units ($e^2 a_0^3$) and $\bar{\Omega}^{(1,1)}$ has units of angstroms squared. Equation (2.6-1) gives the classical-mechanical low-temperature asymptote for the first approximation of the diffusion coefficient, that is, $[\mathcal{D}_{12}]_1$ as $T \rightarrow 0$.

The accuracy of the available London constants is within 5 percent for most gases, and at worst 10 percent for gas pairs containing xenon [34]. By eq (2.6-1) the first approximation for the diffusion coefficient is inversely proportional to the 1/3 power of the London constant; thus the errors in $[\mathcal{D}_{12}]_1$ due to errors in C are less than 4 percent for all gases considered. Numerical values are given in section 5.2.

The valid range of temperature for the low-temperature asymptote is difficult to estimate accurately, but this range may be approximated as follows. First, the upper limit is given by the condition at which the London dispersion energy ceases to dominate interactions. From figure 1 this is estimated to occur at reduced temperatures ≤ 0.2 . Second, the lower limit is determined by the magnitude of quantum effects. These effects depend strongly on the deBoer parameter Λ^* and reduced temperature in a complex manner, and no simple estimate seems possible for the lower limit of temperature for eq (2.6-1). For gas pairs with large values of the deBoer parameter, quantum effects are quite significant at $T^* < 0.2$, as shown in table 1. This suggests that eq (2.6-1) is of only qualitative value for $\Lambda^* > 1$ and $T^* < 0.2$. At $\Lambda^* = 1$ and $T^* \geq 0.1$, eq (2.6-1) is useful only to a 10 percent level of uncertainty. For $\Lambda^* = 0.5$ and $T^* \geq 0.02$ the low-temperature asymptote is accurate to within 3 percent, and for $\Lambda^* < 0.5$ it is even better [43].

b. Diffusion Collision Integrals for Short-Range Interactions

The diffusion collision integral expressions for short-range interactions in terms of the exponential and inverse power models are as follows. The exponential potential, eq (2.3-3), gives

$$\bar{\Omega}^{(1,1)} = 4\pi\alpha^2 \rho^2 I_{(1,1)}, \quad (2.6-2)$$

in which $\alpha = \ln(\varphi_0/kT)$ and $I_{(1,1)}$ is an integral available from tables [35] as a function of α . The inverse power potential, eq (2.3-5), gives

$$\bar{\Omega}^{(1,1)} = \pi \left(\frac{sK}{kT} \right)^{2/s} \Gamma(3-2/s) A^{(1)}(s), \quad (2.6-3)$$

in which $\Gamma(3-2/s)$ is the gamma function of argument $(3-2/s)$ and $A^{(1)}(s)$ is an integral, independent of temperature and available in tables [49, 50] for different values of s .

The reliability of diffusion coefficients calculated from molecular beam experiments is estimated as follows. First, the consistency of \mathcal{D}_{12} by molecular beam results and by direct diffusion experiments can be checked at about 1000 K, a temperature at which these results overlap. The agreement is within a few percent for the gas pairs Hc-Ar, Hc-N₂, and H₂-Ar. Other gas pairs do not have sufficient data for such a comparison. Second, the uncertainties of the potentials themselves as a function of r can be evaluated by comparison (1) with reliable theoretical calculations, (2) with potentials obtained from different apparatus in the same laboratory and from different laboratories, and (3) with potentials derived from other transport property measurements at elevated temperatures [46-48, 51]. The potentials are determined from molecular beam scattering experiments which have been done only at two independent laboratories: Amdur et al., at the Massachusetts Institute of Technology, and Leonas et al., at the Moscow State University. This information has a level of reliability that varies with the type of gas. The noble gas pairs have uncertainties in the potentials that range from about 10 to 30 percent. Gas pairs with diatomic molecules have higher uncertainties, about 20 to 45 percent, and for polyatomic molecules even higher uncertainties, 30 to 60 percent. The diatomic and polyatomic molecules have less reliability than the noble gases because nonspherical characteristics of molecules are not completely taken into account in the derivation of the potential from the experimental scattering observations. In addition, for the dissociated gases H, N, and O, there are only a few molecular beam measurements, which are relatively difficult to obtain; for these mixtures uncertainties in the potentials range from about 30 to 60 percent. However, these rather large uncertainties in the potentials appear only as much smaller uncertainties in the calculated diffusion coefficients. This is clearly evident from eq (2.6-3) for the inverse power potential, since its collision integral is proportional to a fractional power of the potential parameters (the ratio $2/s$ is less than one).

The valid temperature range for diffusion coefficients calculated for short-range interactions can be predicted as follows. The potentials derived from molecular beam scattering experiments are reported with an applicable internuclear separation range. These are obtained directly from the minimum and maximum values of the measured scattering cross sections [46]. In order to calculate the upper and lower limits of the temperature range, the minimum and maximum values of the separation range, respectively, are assumed to be approximately related to the collision integral as $\Omega^{(1,1)} \approx \pi r^2$. Since the collision integral is also given in terms of the potential parameters and temperature by eqs (2.6-2) or (2.6-3), a temperature range can easily be computed. The accuracy of the predicted temperature limits has two significant figures at most.

c. Combination Rules

Often no direct determinations are available for the intermolecular potential of a particular gas

pair, but the potentials for the individual species may be known. Various semi-empirical combination rules are available for the prediction of potential parameters for a 1-2 interaction from those for the 1-1 and 2-2 interactions. Such rules work well enough to allow the prediction of \mathcal{D}_{12} to a level of uncertainty in the order of 10 percent.

The combination rules for the long-range and short-range interactions are as follows. For long-range interactions, theory indicates a geometric-mean rule for the London dispersion coefficient,

$$C_{12} = (C_{11}C_{22})^{1/2}. \quad (2.6-4)$$

This rule has been tested [52] and found to be quite accurate. Theory also suggests, but more weakly, a geometric-mean combination rule for the short-range interactions [53];

Exponential Potential

$$(\varphi_0)_{12} = [(\varphi_0)_{11}(\varphi_0)_{22}]^{1/2}, \quad (2.6-5a)$$

$$\rho_{12}^{-1} = \frac{1}{2}(\rho_{11}^{-1} + \rho_{22}^{-1}), \quad (2.6-5b)$$

and

Inverse-Power Potential

$$K_{12} = (K_{11}K_{22})^{1/2}, \quad (2.6-6a)$$

$$s_{12} = \frac{1}{2}(s_{11} + s_{22}). \quad (2.6-6b)$$

These rules have been directly tested by means of the molecular beam scattering experiments, and the results are quite satisfactory [54-56].

2.7. Determination of Diffusion Coefficients from Other Transport Property Measurements

In this section procedures are described for the determination of \mathcal{D}_{12} from other transport property measurements according to results of the Chapman-Enskog theory [1, 2]. These procedures are virtually independent of knowledge of the molecular interactions, and are an alternate route to the reliable prediction of \mathcal{D}_{12} .

a. Mixture Viscosity

The Chapman-Enskog first approximation for the viscosity of a binary mixture can be expressed [57] as a quadratic equation in the diffusion coefficient:

$$(p\mathcal{D}_{12})^2 a + (p\mathcal{D}_{12}) b + (p\mathcal{D}_{12}) c A_{12}^* + A_{12}^* d = 0, \quad (2.7-1)$$

in which

$$a = (x_1 x_2)^2 (\eta_{\text{mix}} - \eta_1 - \eta_2) / \eta_1 \eta_2, \quad (2.7-2)$$

$$b = 2x_1 x_2 (M_1 + M_2)^{-1} RT [\eta_{\text{mix}} (x_1^2 \eta_2 + x_2^2 \eta_1) - \eta_1 \eta_2] / \eta_1 \eta_2, \quad (2.7-3)$$

$$c = \frac{6}{5} x_1 x_2 (M_1 + M_2)^{-1} RT [\eta_{\text{mix}} (x_1^2 M_1^2 \eta_2 + x_2^2 M_2^2 \eta_1) - (x_1 M_1 - x_2 M_2)^2 \eta_1 \eta_2] / M_1 M_2 \eta_1 \eta_2, \quad (2.7-4)$$

$$d = \frac{3}{5} (2x_1 x_2 RT)^2 \eta_{\text{mix}} / M_1 M_2, \quad (2.7-5)$$

where R is the gas constant ($82.0567 \text{ cm}^3 \cdot \text{atm} / \text{mole} \cdot \text{K}$), η is the viscosity in $\text{g}/\text{cm} \cdot \text{s}$, η_{mix} denotes the mixture viscosity, and the subscripts have their usual meaning. The determination of \mathcal{D}_{12} requires experimental data for mixture composition, the molecular weights and viscosities of the pure components, and the mixture viscosity of the gas pair. The only nonexperimental quantity required is the collision integral ratio A_{12}^* . The variation of A_{12}^* with temperature is only a few percent in the intermediate temperature region, is relatively independent of the choice of a realistic intermolecular potential model, and is insensitive to inelastic collisions (sec. 2.2, part c) and quantum effects (sec. 2.5). Thus the determination of \mathcal{D}_{12} from viscosity measurements essentially eliminates the need for accurate information about molecular interactions.

For a mixture of a gas with itself the binary mixture expression, eq (2.7-1), reduces to

$$p\mathcal{D}_{11} = \frac{6}{5} A_{11}^* (RT/M_1) \eta_1, \quad (2.7-6)$$

in which \mathcal{D}_{11} is known as the self-diffusion coefficient.

The determination of \mathcal{D}_{12} from viscosity measurements has been derived from *first approximation* formulas. On this basis the diffusion coefficients calculated cannot be the true values of \mathcal{D}_{12} , which have a small composition dependence. The diffusion coefficients calculated cannot be exact $[\mathcal{D}_{12}]_1$ because experimental viscosity data are used [58]. However, the diffusion coefficients calculated from experimental binary mixture viscosity data are nearly equal to \mathcal{D}_{12} at a mixture composition corresponding to the heavy component in trace amounts, as shown by numerical computations of the higher Chapman-Enskog approximations [10]. The uncertainty in this conclusion was found to be less than any error in available diffusion coefficient measurements.

The reliability of \mathcal{D}_{12} calculated from mixture viscosity measurements is almost the same as obtainable by \mathcal{D}_{12} measurements with the best modern techniques, as shown by the following analysis. First, assume that A_{12}^* is known exactly. On the basis of an error propagation analysis of eq (2.7-1), the calculation procedure for \mathcal{D}_{12} can introduce a loss in precision by as much as a factor of five [57]. However, reliable viscosity measurements are obtained with uncertainties of 1/10 percent at about room temperature and about 1/2 percent at 1000 K. These uncertainties are approximately 10 times less than in direct \mathcal{D}_{12} measurements at the corresponding temperatures. Second, remove the restriction of a perfectly known A_{12}^* in order to obtain the total uncertainty of calculated \mathcal{D}_{12} . For spherical or homonuclear diatomic molecules at intermediate temperatures A_{12}^* is reliable to about 1 percent; nonspherical or polar gases have slightly larger uncertainties in A_{12}^* . Uncertainties in values of A_{12}^* will be directly reflected in \mathcal{D}_{12} , that is, a 1 percent error in A_{12}^* corresponds to an error of approximately 1 percent in \mathcal{D}_{12} . Thus the total uncertainty in diffusion coefficients calculated from accurate viscosity

measurements is about 2 percent at room temperature, an uncertainty comparable to the available direct \mathcal{D}_{12} measurements.

b. Thermal Conductivity

The first approximation of the Chapman-Enskog theory for the thermal conductivity of binary mixtures can be used to compute values of \mathcal{D}_{12} [59]. The procedure is similar to that used for diffusion coefficients calculated from viscosity data, but the values calculated from thermal conductivity measurements are not as reliable as available \mathcal{D}_{12} measurements for two reasons. First, the relationship between thermal conductivity and \mathcal{D}_{12} is slightly more sensitive to temperature and molecular interactions; that is, the applicable relationship has the collision integral ratio B_{12}^* , as well as A_{12}^* . Second, the accuracy of thermal conductivity data is only equal to, and often less than, that of \mathcal{D}_{12} measurements, and the experimental errors propagate by a factor of as much as five through these calculations. Thus thermal conductivity is a transport property from which only mediocre estimates of \mathcal{D}_{12} are possible at present. Moreover, except for the rare gases, thermal conductivity also depends on the molecular internal degrees of freedom.

An alternative approach is to calculate B_{12}^* from λ_{mix} and a known value of \mathcal{D}_{12} at the same temperature (the value of A_{12}^* is still assigned theoretically). Since B_{12}^* is related to the temperature derivative of \mathcal{D}_{12} , the temperature range of \mathcal{D}_{12} can be extended. That is, if λ_{mix} and \mathcal{D}_{12} are known at a single temperature, values of \mathcal{D}_{12} can be predicted at nearby temperatures [59].

c. Thermal Diffusion Factor

The Chapman-Enskog theoretical first approximation for the thermal diffusion factor of binary mixtures may give reliable values of \mathcal{D}_{12} . The thermal diffusion factor describes how a gas mixture separates under the influence of a temperature gradient. Diffusion coefficients can be calculated from the strong composition dependence of the thermal diffusion factor, α_T [60]. But, the available measurements of the composition dependence of α_T have rather large uncertainties, which lead to mediocre values of \mathcal{D}_{12} at present. Another procedure relates the temperature dependence of \mathcal{D}_{12} to that of α_T , and the derived relationship is combined with a single measurement of \mathcal{D}_{12} to produce diffusion coefficients over a wide temperature range [61]. From this procedure the accuracy of \mathcal{D}_{12} is good, because uncertainties in the measurements appear only as much smaller uncertainties in the calculated diffusion coefficients. In principle the calculations are applicable generally, but have been limited to gas pairs with $M_2/M_1 \ll 1$ and a trace concentration of the heavy component. The procedure has involved iterative type calculations which are described next.

An "experimental" value of $(6C_{12}^* - 5)$ is compared to the auxiliary theoretical expression

$$(6C_{12}^* - 5) = 2[2 - (\partial \ln [\mathcal{D}_{12}]_1 / \partial \ln T)_P], \quad (2.7-7)$$

in which the "experimental" $(6C_{12}^* - 5)$ is derived as follows:

$$(6C_{12}^* - 5) = \alpha_T [(1 + \kappa_2) (-S_2/Q_2)]^{-1}, \quad (2.7-8)$$

$$-S_2 = \frac{15 M_1 (M_1 - M_2)}{2 (M_1 + M_2)^2} + 4 \frac{M_1 M_2}{(M_1 + M_2)^2} A_{12}^* \\ - \frac{5 M_2^2}{3 (M_1 + M_2)} \frac{p [\mathcal{D}_{12}]_1}{[\eta_2]_1 RT}, \quad (2.7-9)$$

$$Q_2 = \frac{10}{3} \frac{M_2}{(M_1 + M_2)^2} \frac{p [\mathcal{D}_{12}]_1}{[\eta_2]_1 RT} (3M_1^2 \\ + M_2^2 + \frac{8}{5} M_1 M_2 A_{12}^*), \quad (2.7-10)$$

$$\kappa_2 = \frac{1}{42} (8E_{22}^* - 7)^2 + \frac{2}{21} \left(1 - \frac{M_2}{M_1}\right) (8E_{22}^* - 7) \\ \left[1 - \frac{3}{4} (5 - 4B_{12}^*) (6C_{12}^* - 5)^{-1}\right], \quad (2.7-11)$$

$$8E_{22}^* - 7 \approx 2[1 - (\partial \ln \eta_2 / \partial \ln T)_p]. \quad (2.7-12)$$

In these equations the subscript 2 denotes the light component and 1 the heavy, κ_2 is a small correction term, and for Q_2 the Kihara expression is used in this case. As previously discussed, A_{12}^* is virtually independent of temperature and the potential model. The values of $[\mathcal{D}_{12}]_1$ and $[\eta_2]_1$ can be interpreted as "experimental first approximations." In eq (2.7-8) the denominator is weakly dependent on temperature, but the major temperature dependence is in α_T , and this is obtained from experiments. The substitution of eqs (2.7-9) to (2.7-12) into the right-hand side of eq (2.7-8) gives the "experimental" $(6C_{12}^* - 5)$ value principally in terms of the temperature dependence of α_T . For the first interaction step $(-S_2/Q_2)$ and κ_2 are assumed independent of temperature, the temperature at which they are evaluated is conveniently taken to be the same as for the experimental \mathcal{D}_{12} . This value of $(6C_{12}^* - 5)$ is substituted into the differential equation eq (2.7-7), and the subsequent integration completes the first iteration cycle. The constant of integration is evaluated from one isothermal measurement of \mathcal{D}_{12} . The result is a relationship for the temperature dependence of \mathcal{D}_{12} over the range for which measurements of α_T are available. The second iteration step uses values of $[\mathcal{D}_{12}]_1$ from the first cycle together with experimental values of $[\eta_2]_1$ to evaluate the temperature variation of $(-S_2/Q_2)$; κ_2 can be assumed independent of temperature. The second set of $(-S_2/Q_2)$ gives new values for $(6C_{12}^* - 5)$, and new values of $[\mathcal{D}_{12}]_1$ by the integration of eq (2.7-7). The $[\mathcal{D}_{12}]_1$ of the second set are usually almost identical with the first set, but a third iteration step can be used as a check, if desired. The diffusion coefficients calculated are as reliable as most direct measurements of \mathcal{D}_{12} ; at present this means about a few percent.

In some cases the calculation procedure can be simplified, and made to involve the thermal diffusion factor in a more direct way [62], but this method was not used for any results in this report.

The determination of \mathcal{D}_{12} from thermal diffusion data is, strictly speaking, limited to noble gas pairs.

The equations are based on monatomic molecules which are free of internal energy. The theoretical expressions can be used for polyatomic gases when the translational energy contribution is much greater than that of internal energy factors which contribute to α_T .

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3. Experimental Methods for Diffusion Coefficients

The purpose of this section is to assess reliabilities and limitations of the various experimental methods used for determinations of gaseous diffusion coefficients. The critical evaluation of \mathcal{D}_{12} necessarily requires a comprehensive appraisal of experimental methods, which has not been prepared before. The various methods are outlined in section 3.1 in order to give an overall perspective of the types of apparatus and the reliabilities of results. In section 3.2 five major methods are described and their systematic errors considered. A major method means one that has been frequently used by different investigators and has well-known experimental uncertainties. The results of four of these five methods have generally contributed data of high quality, but one technique—the evaporation-tube—has had disappointing results and is included as a major method only because it has been used more often than any other. In section 3.3 brief descriptions are given for six methods which have not been used very often, but which have sufficient results available so that their reliability may be estimated. These are called minor methods and the results have made small contributions to the recommended values. The final section of this chapter contains remarks about seven miscellaneous methods which have not contributed to the recommended values, but which are of general applicability or of unusual inventiveness. Every experimental method ever used is not included in these groups, but those omitted are considered unimportant.

The discussion of each method includes a comprehensive list of references to specific studies; these listings contain the reference information for the discussion of each method unless special footnotes are given in the text.

Several of the experimental methods have been previously described in specialized surveys [1-6].³

The first significant measurements of diffusion in gases were made by Thomas Graham, starting in 1829. His ingenious experimentation included observations of gaseous diffusion in closed-tube and two-bulb apparatuses [7, 8]. These techniques were later developed into the most reliable methods, by modern standards, for the determination of dif-

fusion coefficients. In addition Graham used what is now called the capillary-leak method and an equivalent to the diffusion bridge. However, Graham never calculated a diffusion coefficient, and actually most of his work preceded the mathematical statement of the law of diffusion by Fick in 1855 [9]. From some of Graham's later observations, reported in 1863, the first accurate \mathcal{D}_{12} were calculated by Maxwell in 1867 [10, 11]. Until recent times most of Graham's work in diffusion had been overlooked [11].

In the 1870's two experimental methods, the closed tube and the evaporation tube, were devel-

TABLE 2. Classification of experimental methods

Name	Primary Investigator(s)	Reliability
Major		
Closed Tube.....	Loschmidt (1870 a, b).....	Good.
Evaporation Tube.....	Stefan (1873).....	Poor.
Two-Bulb Apparatus.....	Ney and Armistead (1947).....	Good.
Point Source.....	Walker and Westenberg (1958 a, b).....	Average.
Gas Chromatography....	Giddings and Seager (1960). ^a	Average.
Minor		
Open Tube.....	von Obermayer (1882 a); Waitz (1882 a, b).....	Average.
Back Diffusion.....	Harteck and Schmidt (1933).....	Average.
Capillary Leak.....	Klibanova et al. (1942).....	Poor.
Unsteady Evaporation.....	Arnold (1944).....	Fair.
Diffusion Bridge.....	Bendt (1958).....	Average.
Dissociated Gases.....	Wise (1959); Kronigell and Strandberg (1959).....	Poor.
Miscellaneous		
Droplet Evaporation....	Langmuir (1918); Katan (1969).....	?
Dufour Effect.....	Waldmann (1944).....	?
Thermal Separation Rate.....	Nettley (1954).....	?
Kirkendall Effect.....	McCarty and Mason (1960).....	?
Sound Absorption.....	Holmes and Tempest (1960).....	?
Cataphoresis.....	Hogervorst and Freudenthal (1967).....	?
Resonance Methods.....	See text.....	?

^a In 1960 four independent gas chromatography studies were submitted for publication; for details see section 3.2, part d.

³ Figures in brackets indicate the literature references at the end of Section 3.

oped; their results include almost all values of \mathcal{D}_{12} up until World War II. Then several other methods (two-bulb apparatus, point source, diffusion bridge, dissociated gases, and gas chromatography) were developed because of interests in isotope separations, combustion processes, and theoretical studies of intermolecular forces which were in need of values of \mathcal{D}_{12} over an extensive range of temperatures. The availability of radioisotopes made measurements for many gas pairs easier. In addition a number of other techniques have been occasionally used over the last half century. The experimental methods are classified in table 2, and the reference sources can be found in Bibliography I.

3.1. Outline of Experimental Methods

Table 2 serves as an outline of the assessment of methods that follows. The reliabilities given are based on reproducibilities and on intercomparisons of \mathcal{D}_{12} by various methods. At present the reliability [12] is not exactly known for each method; these measurement techniques are amenable to possible refinements. The determinations of \mathcal{D}_{12} are considered good when uncertainties are within about 2 percent, although for a given apparatus the reproducibility of results may be better than 1 percent. A vast majority of available data does not have this level of either reproducibility or reliability. Determinations of \mathcal{D}_{12} are considered of average quality when uncertainties are within about 5 percent. These magnitudes indicate that accurate determinations of diffusion coefficients are rather difficult, even with the best of modern instrumentation.

The major and a few minor methods are schematically illustrated in figure 2, classified according to overall geometry of apparatus and time behavior of the diffusion process. The apparatus listed under the first two columns have no carrier gas flow in the zone where diffusion takes place. The two apparatus in the third column have diffusion occurring within a flowing gas stream.

3.2 Major Experimental Methods

a. Closed Tube

In 1870 the closed-tube method was developed by Loschmidt, who carefully determined \mathcal{D}_{12} for 10 gas pairs at temperatures of 252 to 293 K. The essential characteristic of this method is a variation of mixture composition with time and position throughout a long tube closed at both ends. The gases of the mixture are initially separate in the closed tube, then interdiffuse at constant temperature and pressure. The diffusion time is controlled by an opening mechanism at the middle of the tube. The composition changes are measured as a function of time, either continuously or after a definite period of diffusion.

Determinations of \mathcal{D}_{12} by the closed-tube method are usually quite reliable. The results have been obtained at temperatures from 195 to 478 K. This range indicates an indirect disadvantage—determinations at more extreme temperatures have not been made because of difficulties that arise from the construction and the operation of a thermostat around a long tube (about 1 meter) with moving parts.

The reported determinations are listed in table 3 in chronological order. There are various versions

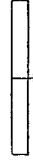


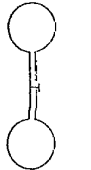

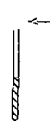
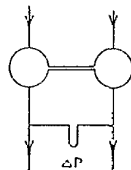
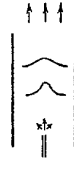
TYPE TIME BEHAVIOR	TWO-SIDED	ONE-SIDED	FLOW
UNSTEADY	 CLOSED TUBE	 OPEN TUBE	 PULSE SOURCE (GAS CHROMA- TOGRAPHY)
QUASI- STEADY	 TWO-BULB	1.  2.  1. CAPILLARY LEAK 2. EVAPORATION TUBE	
STEADY	 DIFFUSION BRIDGE		 POINT SOURCE

FIGURE 2. Principal experimental methods for diffusion coefficients.

of the closed-tube apparatus, but details of these refinements are omitted here.

The basis of all closed-tube determinations is a solution of the one-dimensional time-dependent diffusion equation,

$$\partial x_1 / \partial t = \mathcal{D}_{12} (\partial^2 x_1 / \partial z^2), \quad (3.2-1)$$

where \mathcal{D}_{12} has been assumed independent of mixture composition and position. In eq (3.2-1) x_1 is the mole fraction of component 1 in the binary mixture, and t and z are time and axial distance, respectively. For the initial and boundary conditions,

$$\begin{aligned} x_1 &= x_1^I & 0 \leq z < L/2, t = 0 \\ x_1 &= x_1^II & L/2 < z < L, t = 0 \\ \partial x_1 / \partial z &= 0 & z = 0 \text{ and } z = L, t \geq 0, \end{aligned}$$

the solution of eq (3.2-1) is

$$x_1(z, t) = \frac{1}{2} (x_1^I + x_1^II) + \frac{2(x_1^I - x_1^II)}{\pi} \sum_{n=0}^{\infty} \frac{e^{-(2n+1)^2 t / \tau}}{(2n+1)} \sin \frac{(2n+1)\pi z}{L}, \quad (3.2-2)$$

and the relaxation time is

$$\tau = L^2 / \pi^2 \mathcal{D}_{12}, \quad (3.2-3)$$

TABLE 3. Determinations of \mathcal{D}_{12} by the closed-tube method^a

Author(s)	Date	Author(s)	Date
Loschmidt.....	1870 a, b	Amdur and Schatzki.....	1957; '58
Wretschko.....	1870	Carswell.....	1960
von Obermayer.....	1880, '82 b, '83, '87	Suetin et al.....	1960
Rutherford and Brooks.....	1901	Suetin and Ivakin.....	1961
Schmidt.....	1904	Amdur and Shuler.....	1963
Lonius.....	1909	Carswell and Stryland.....	1963
Wintergerst.....	1930	Holsen and Strunk.....	1964
Harteck and Schmidt.....	1933	Ivakin and Suetin.....	1964 a, b
Boardman and Wild.....	1937	Suetin.....	1964
Coward and Georgeson.....	1937	Amdur and Beatty.....	1965
Hirst and Harrison.....	1939	Amdur and Malinauskas.....	1965
Braune and Zehle.....	1941	Cordes and Kerl.....	1965
Croth and Harteck.....	1941	Ljunggren.....	1965
Heath et al.....	1941	Reichenbacher et al.....	1965
Croth and Sussner.....	1944	Fedorov et al.....	1966
Wall and Kidder.....	1946	Kosov and Abdullina.....	1966
Hutchinson.....	1947	Arnold and Toor.....	1967
Boyd et al.....	1951	Gover.....	1967
Timmerhaus and Drickamer.....	1951	Manner.....	1967
Amdur et al.....	1952	Ivakin et al.....	1968
Strehlow.....	1953	Beatty.....	1969
Bunde.....	1955	Baker.....	1970 a
Rumpel.....	1955	Belousova et al.....	1970
		Jacobs et al.....	1970

^a Complete reference information is given in Bibliography I.

where L is the total length of the closed tube. A few additional assumptions were taken to obtain eq (3.2-2), namely a uniform cross section and symmetry about the midplane at $L/2$. Equation (3.2-2) may be simplified in accordance with the technique used for composition analysis; the simplified expressions are readily available elsewhere [3, 6, 13, 14], as well as from the original articles of table 3.

The determination of \mathcal{D}_{12} requires measurements of composition, temperature, pressure, geometrical factors, and time. The uncertainties of these measurements are usually much less than the reliability of the results; thus the reliability is apparently dependent on other factors. Occasionally, however, poor methods of composition analyses have led to inaccurate results.

Other possible uncertainties of the closed-tube method are as follows: Errors due to convective mass flux are possible. To avoid convection from buoyancy effects, the lighter gas should always be placed in the top half of vertically mounted apparatus. If the closed tube is in a horizontal orientation, a "spillage" convective flux may occur; that is, the higher density component in one half may spill across the diffusion "interface" into the lower portion of the other half, and the low density component would then flow into the upper portion of the opposite half of the closed tube. Spillage is not significant if the diffusing component is a tracer. At the start of diffusion, convection effects are also possible because of the movements of the opening mechanism; this has been investigated [15-17], and the reproducibility of results from run to run indicates that the effect is small. Convection effects are also possible because of nonuniform temperatures axially along the tube. The design and operation of closed-tube apparatus should eliminate all possible convection effects.

A significant uncertainty, even though not limited to the closed-tube method, is the small dependence of \mathcal{D}_{12} on the mixture composition. The diffusion coefficient was assumed independent of mixture composition. Equation (3.2-1) rewritten to express

the composition dependence of \mathcal{D}_{12} is

$$\partial x_1 / \partial t = \mathcal{D}_{12} (\partial^2 x_1 / \partial z^2) \\ + (\partial x_1 / \partial z)^2 (\partial \mathcal{D}_{12} / \partial x_1). \quad (3.2-4)$$

If one of the components is a tracer then the composition gradient, $\partial x_1 / \partial z$, is very small, and the uncertainty essentially zero. If two pure gases fill each half of the closed tube, then both the gradient or the composition dependence of \mathcal{D}_{12} may be significant. However, for the case of the closed tube, the exact integration of the diffusion equation with a composition-dependent \mathcal{D}_{12} has not been performed. The uncertainty of results caused by the composition dependence of \mathcal{D}_{12} depends on the duration of the experimental run, the gas pair investigated, and the initial composition of the mixture. Calculated values of \mathcal{D}_{12} would apparently depend on the length of the experimental run. This has been investigated [15, 18], and the variation of \mathcal{D}_{12} is small for diffusion times between 10 min and about 2 hrs. To a first approximation the composition of the mixture may be taken equal to a uniform mixture of the components, or the arithmetic mean of the initial conditions [17].

The uncertainty in determinations of \mathcal{D}_{12} caused by the assumption of a one-dimensional diffusion equation has not been estimated.

Additional uncertainties may be caused by the Dufour effect. The Dufour effect is a small temperature transient that occurs when two gases interdiffuse, and may occur even with ideal gases. The uncertainty in \mathcal{D}_{12} caused by the Dufour effect can be made small by suitable choice of apparatus geometry [17] but most experimenters have apparently simply ignored the problem. If the mixture were nonideal then diffusion would be accompanied by heats of mixing or pressure changes.

The closed-tube determinations of \mathcal{D}_{12} often have reproducibilities better than 1 percent, and the measurements have been reported accurate to 1 to 3 percent. However, independent determinations of \mathcal{D}_{12} for the same gas pair indicate that this

method has a reliability no better than 2 percent. These comparisons are presented in deviation plots given in section 5.3. To achieve greater reliability a major effort involving careful variation of many experimental parameters would probably be necessary.

b. Two-Bulb Apparatus

The two-bulb method was developed by Ney and Armistead in order to determine the self-diffusion coefficient of UF_6 ; their results were published in 1947. Two bulbs, or chambers, are connected by a narrow tube through which the diffusion occurs. After an initial transient, the composition in the bulbs varies exponentially with time, and \mathcal{D}_{12} can be found from the relaxation time.

The determinations of \mathcal{D}_{12} by the two-bulb method have been made over a temperature range of 65 to 400 K, with one datum available at 473 K. This range of temperatures is ~ 50 K larger than results obtained by the closed-tube method. In general, measurements at different temperatures are easier to make with the two-bulb apparatus because its relative compactness facilitates thermostating, and its opening mechanism can be designed without moving parts. These conveniences, however, are only operational advantages; the ultimate accuracies of \mathcal{D}_{12} are probably the same as determined by both the closed-tube and two-bulb methods.

Studies by two-bulb apparatus are listed in chronological order in table 4. This listing shows that the method has been widely used in recent years. The meticulous studies by van Heijningen et al. are especially noteworthy because the results attained are probably the most reliable measurements of \mathcal{D}_{12} to date, within 1 percent, over a temperature range of 65 to 400 K.

As for the closed tube, the simple theory for the two-bulb apparatus involves the assumptions of constant pressure and temperature, constant \mathcal{D}_{12} , and one-dimensional diffusion. It also involves the following additional assumptions:

(1) Quasi-stationary state—the flux of a component is constant along the connecting tube. Since \mathcal{D}_{12} is assumed constant, this implies a linear variation in composition in the tube.

(2) The connecting tube volume is much smaller than either bulb volume. This is related to the quasi-stationary-state assumption.

(3) The composition gradient is entirely contained in the connecting tube.

With all these assumptions introduced into the diffusion equation for one component, eq (2.1-1), a simple solution can be obtained of the form

$$\Delta x(t) = \Delta x(0) \exp(-t/\tau), \quad (3.2-5)$$

where $\Delta x(t)$ is the composition difference at $t = \infty$ and at time t in one bulb, $\Delta x(0)$ is the composition difference between $t = \infty$ and $t = 0$, and τ is the relaxation time. The component subscript 1 has been dropped from the notation. The relaxation time is

$$\tau = \frac{1}{\mathcal{D}_{12}} \left(\frac{L}{A} \right) \left(\frac{V_1 V_2}{V_1 + V_2} \right), \quad (3.2-6)$$

where A is the cross-sectional area, L the length of the tube connecting the bulbs, and V_1 and V_2 denote the bulb volumes. From measurements as a function of time of the composition in one bulb, or alternately the composition difference between the two bulbs, the relaxation time is obtained from eq (3.2-5). Corrections to the relaxation time for the above assumptions are as follows.

TABLE 4. Determinations of \mathcal{D}_{12} by the two-bulb method^a

Author(s)	Date	Author(s)	Date
Ney and Armistead.....	1947	Mueller and Cahill.....	1964
Winn and Ney.....	1947	Watts.....	1964
Winn.....	1948	Brown and Murphy.....	1965
Hutchinson.....	1949	Malinauskas.....	1965
Winn.....	1950	Mason et al.....	1965
Schäfer et al.....	1951	Watts.....	1965
Visner.....	1951 a, b	Chakraborti and Gray.....	1966
Winter.....	1951	Kosov and Novosad.....	1966 a
DeLuca.....	1954	Malinauskas.....	1966
Schäfer and Moesta.....	1954	Paul and Watson.....	1966
Andrew.....	1955	Saran and Singh.....	1966
Schäfer and Schuhmann.....	1957	Srivastava and Saran.....	1966 a, b
Saxena and Mason.....	1959	van Heijningen et al.....	1966
Schäfer.....	1959	Vužić and Milojević.....	1966
Srivastava and Srivastava.....	1959	Oost et al.....	1967
Srivastava.....	1959	Singh et al.....	1967
Srivastava and Barua.....	1959	Annis et al.....	1968
Miller and Carman.....	1961	Malinauskas.....	1968
Paul and Srivastava.....	1961 a, b, c	Mathur and Saxena.....	1968
Weissman et al.....	1961	Singh and Srivastava.....	1968
Zmbov and Knežević.....	1961	van Heijningen et al.....	1968
Durbin and Kobayashi.....	1962	Annis et al.....	1969
Paul.....	1962	DuBro.....	1969
Srivastava and Paul.....	1962	Malinauskas and Silverman.....	1969
Srivastava and Srivastava.....	1962	Vugts et al.....	1969
Srivastava.....	1962	Weissman.....	1969
Golubev and Bondarenko.....	1963	DuBro and Weissman.....	1970
Schäfer and Reinhard.....	1963	Humphreys and Mason.....	1970
Srivastava and Srivastava.....	1963	Lannus and Grossmann.....	1970 a, b
Wendt et al.....	1963	Mistler et al.....	1970
Bondarenko and Golubev.....	1964	Vugts et al.....	1970
Mason et al.....	1964 a, b	Weissman and DuBro.....	1970 a, b
Miller and Carman.....	1964	Vugts et al.....	1971

^a Complete reference information is given in Bibliography I.

The quasi-stationary state assumption is unnecessarily severe [19]; it is sufficient to assume only that the mean flux in the tube is proportional to the effective mean flux at the two ends of the tubes. On this basis a correction factor K for the relaxation time may be derived,

$$\tau = \frac{K}{\mathcal{D}_{12}} \left(\frac{L}{A} \right) \left(\frac{V_1 V_2}{V_1 + V_2} \right), \quad (3.2-7)$$

where

$$K = 1 + \frac{AL}{3V_1} \left(\frac{1 - \beta + \beta^2}{1 + \beta} \right), \quad (3.2-8)$$

with $\beta = V_1/V_2$. This approximate solution assumes that the composition analysis is performed in bulb V_1 and that AL/V_1 is small. Deviations from a quasi-stationary state show up as values of K unequal to unity. The deviations from the quasi-stationary state are due to the fact that the bulbs are not infinitely large compared to the connecting tube. To minimize these deviations the apparatus should be constructed such that the volume of the tube is very much less than the volume of either bulb. For bulbs of equal size $K = 1 + AL/6V$, where V is the volume of a bulb.

Transient effects arise from the finite time required to establish a constant gradient across the entire length of the tube [20]. These transients can be avoided by waiting for some time to elapse after the start of mixing. Normally, the transients decay rapidly, and they completely disappear within a few minutes.

The assumption that the composition gradient is all in the connecting tube requires an end correction. This correction is required because the gradient does not truncate immediately at either outlet of the tube. The extension of the gradient into the bulbs is corrected for by a small increase in the actual length of the tube. The end correction is given by

$$L_{\text{eff}} = L + 2\alpha R, \quad (3.2-9)$$

where L_{eff} is the effective length of the tube, R is its radius, α is a numerical constant whose value depends on the geometrical configuration of the end of the tube, and the factor 2 accounts for both ends of the tube. The value of α is obtained from an analogous case for sound passage in a tube [21, 22]. Typical values of α are as follows: $\alpha = 0.58$ when the connecting tube end is in free space, $\alpha = 0.82$ when the end is flush with a flat surface, and $\alpha \geq 0.82$ when the end is flush with the inside surface of a spherical bulb. The value of $\alpha = 0.82$ has been invariably used in the analysis of two-bulb apparatus experiments, but this is not always correct. The choice of an incorrect α value has led, in a few studies, to systematic errors of the order of 1 percent.

Most two-bulb apparatus are constructed with the connecting tube of uniform bore; if not so, then the ratio L/A is taken to mean $\sum_j (L_j/A_j)$ for each element of length L_j and cross section A_j [23].

In addition to the above corrections, observations may require corrections for Knudsen flow [24, 25], which can occur during diffusion in narrow capillaries at low pressures, where the mean free path is not negligible compared to the diameter of the connecting tube.

The determinations of \mathcal{D}_{12} by the two-bulb method have uncertainties similar to those of the closed tube, but with less chance of convective effects because of the narrowness of the connecting tube. In several two-bulb investigations, errors from non-negligible sample volumes are possible because samples of the mixture were removed from the apparatus during the diffusion run. The inaccuracies of two-bulb measurements have been reported to be between 1 and 3.5 percent. Except for results by van Heijningen et al. the reliability of data by this method is considered no better than 2 percent. This is the same as for the closed tube. The two-bulb method is capable of yielding reliable \mathcal{D}_{12} provided care is taken to optimize the geometry of the apparatus and to make corrections.

c. Point Source

The point-source method was developed especially for the determination of diffusion coefficients at high temperatures. In 1958 Walker and Westenberg fully reported the first results by this method, in which a trace amount of gas is steadily introduced through a fine hypodermic tube into a carrier gas flowing in the same direction. The tracer spreads by diffusion through the carrier gas, which has characteristics of steady-state laminar flow with a flat velocity profile. The mixture composition is measured by means of a sample probe located at various distances downstream of the tracer inlet.

Point source determinations of \mathcal{D}_{12} are available from room temperature up to 1944 K.

Studies by this method are listed in chronological order in table 5. A few special remarks are as follows. Walker and Westenberg used electrical heat and attained temperatures up to ~ 1200 K; Ferron et al. used combustion heat (mixtures containing H_2O or CO_2 were studied) and reached higher temperatures, up to 1944 K.

TABLE 5. Determinations of \mathcal{D}_{12} by the point-source method^a

Author(s)	Date
Westenberg and Walker ^b	1957
Walker.....	1958
Walker and Westenberg.....	1958 a, b, '59, '60
Walker et al.....	1960
Westenberg and Frazier.....	1962
Ember et al.....	1962, '64
Pakurar and Ferron.....	1964, '65, '66
Pakurar.....	1965
Walker and Westenberg.....	1966
Ferron.....	1967
Walker and Westenberg.....	1968

^a Complete reference information is given in Bibliography I.

^b Preliminary note about the development of the point-source method.

The basic equation for the point-source method is

$$\mathcal{D}_{12} \left[\frac{\partial^2 x}{\partial z^2} + \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial x}{\partial r} \right) \right] - U(r) \frac{\partial x}{\partial z} = 0, \quad (3.2-10)$$

where x denotes the tracer component, U is the carrier velocity, z is the axial distance from the point of injection, and r is the radial coordinate measured from the axis. The appropriate boundary conditions are:

$$\lim_{R \rightarrow \infty} x = 0, \\ (\partial x / \partial r)_{r=0} = 0,$$

and the normalization condition

$$Q = \lim_{R \rightarrow 0} 4\pi R^2 \mathcal{D}_{12} (\partial x / \partial R),$$

where $R^2 = r^2 + z^2$ and Q is the volumetric flow rate of the tracer. Equation (3.2-10) involves the usual assumptions of constant temperature and pressure, and the important assumption that the tracer is present at a vanishingly dilute concentration so that the density may be taken constant everywhere. This assumption assures the composition-independence of \mathcal{D}_{12} . Additional assumptions are as follows:

- (1) Steady-state conditions of flow.
- (2) Axial symmetry of the tracer concentration in the carrier gas.
- (3) The flow velocity is uniform everywhere in the field.
- (4) Absence of convection effects.
- (5) Absence of chemical reactions. This assumption is particularly noted, even though applicable to all major methods, because point-source measurements at high temperatures may be accompanied by reactions between the components or the components and surfaces of the diffusion apparatus.

The solution of eq (3.2-10) is

$$x = (Q/4\pi R \mathcal{D}_{12}) \exp [-(R-z)U/2\mathcal{D}_{12}]. \quad (3.2-11)$$

A concentration profile can be used to determine \mathcal{D}_{12} , that is, at a fixed axial distance z , samples of the stream are taken as a function of R . A plot of $\ln(xR)$ versus $(R-z)$ should be linear with slope $-U/2\mathcal{D}_{12}$, and measurements of the carrier velocity U lead to values of \mathcal{D}_{12} . Alternately, the stream can be sampled at points on the axis, $z = R$, and values of \mathcal{D}_{12} calculated from

$$\mathcal{D}_{12} = Q/4\pi(x)_{\max}z, \quad (3.2-12)$$

where $(x)_{\max}$ is the concentration of the tracer at points on the z axis. This axial decay method has the advantage of not requiring knowledge of U , and of requiring significantly fewer composition measurements. The more difficult concentration profile method can serve as a check on the consistency of both the theory of the experiment and the results.

The instrumentation errors of the point-source method have been estimated by Ferron et al. to be about 5 percent. This estimate was based on an approximate analysis of errors arising from measurements of flow, sample probe position, composition, and temperature. These experiments are probably not as reproducible as those by Walker and Westenberg, who performed experiments at lower temperatures and with a more precise technique for composition analysis.

In addition to the instrumentation errors, the possible causes of uncertainty for the point-source method are:

- (1) The unavoidable wake caused by the injector tube for the tracer.
- (2) Difference in density between the tracer and the carrier.
- (3) Variations in the steady-state flow rate of the tracer, or carrier.
- (4) Skewness of the mixture velocity profile.
- (5) Temperature gradients in the stream.

A priori estimates of uncertainties caused by such

effects are difficult to make; but these effects have been empirically investigated [26, 27]. The reliability of point-source measurements of \mathcal{D}_{12} is best estimated by comparisons with the results of other methods. Such comparisons show deviations of up to 4 percent for 10 gas pairs at about 300 K. The deviations are slightly greater at 1000 K by comparison of point-source results and those calculated from short-range interaction forces obtained by molecular-beam scattering experiments (sec. 2.6, part b). The general reliability of \mathcal{D}_{12} by the point-source method is considered to be better than 5 percent, or average.

d. Gas Chromatography

The gas-chromatography method is a flow method in which a trace amount of gas is injected as a pulse into a carrier gas flowing through a long hollow tube. The dispersion of the pulse is caused by the combined action of molecular diffusion and the parabolic velocity profile of the carrier gas. As the pulse emerges from the tube outlet, measurements of the dispersion—characterized by a Gaussian distribution function—lead to values of \mathcal{D}_{12} .

The advantages of the gas-chromatography method are as follows. Determinations of \mathcal{D}_{12} can be completed in a matter of minutes and vapor-gas mixtures can be studied. Once the carrier gas is at temperature and pressure, the injection of a number of sample pulses into the gas is possible, with the result that several samples may be simultaneously dispersing in the tube. The dispersion characteristics of the pulse can be obtained by one simple measurement of its variance. A vapor-gas determination of \mathcal{D}_{12} is practical because of the small amount of sample required to make a pulse. These advantages are operational, only.

Determinations of \mathcal{D}_{12} by gas chromatography are available between temperatures of 77 and 523 K. These studies are listed in chronological order in table 6, all of which are based on the instrumentation

TABLE 6. Determinations of \mathcal{D}_{12} by the gas-chromatography method^a

Author(s)	Date
Giddings and Seager.....	1960
Bohemen and Purnell.....	1961
Bournia et al.....	1961
Fejes and Czárán.....	1961
Giddings and Seager.....	1962
Knox and McLaren.....	1963
Seager et al.....	1963
Barr and Sawyer.....	1964
Knox and McLaren.....	1964
Evans and Kenney.....	1965
Fuller and Giddings.....	1965
Huber and van Vught.....	1965
Chang.....	1966
Arai et al.....	1967
Arnikar et al. ^b	1967 a, b
Fuller and Giddings.....	1967
Giddings.....	1967
Hargrove and Sawyer.....	1967
Giddings.....	1968
Huang et al.....	1968
Zhukhovitskii et al.....	1968
Arnikar and Ghule.....	1969
Fuller et al.....	1969
Wasik and McCulloh.....	1969
Hu and Kobayashi.....	1970
Nagata and Hasegawa.....	1970

^a Complete reference information is given in Bibliography I.
^b Packed chromatography column.

and certain aspects of the conventional theory of gas chromatography. In 1960 four independent manuscripts, which described the method, were submitted for publication: Bohemen and Purnell (23 June); Fejes and Czárán (20 July); Giddings and Seager (3 August), and Bournia, Coull, and Houghton (8 November). Of these authors, only Giddings et al. have continued to publish new determinations of \mathcal{D}_{12} .

Packed chromatography columns have been used, on occasion, to determine \mathcal{D}_{12} . A packed column has a complex geometry because of the interstitial flow volume. Since the geometry and the pulse velocity profile are not well defined in packed columns, their use for determinations of absolute values of \mathcal{D}_{12} is rather uncertain [28].

Outside the scope of this report, but worthy of mention, is the following. Gas chromatography is well suited for high-pressure determinations of \mathcal{D}_{12} because the column actually consists of small-bore tubing which is easily pressurized. High-pressure studies in other apparatus ordinarily require the fabrication of relatively expensive pressure vessels.

Before gas-chromatography apparatus was applied to the determinations of \mathcal{D}_{12} , the theory had been developed for diffusion phenomena in the flow of fluids [29-34]. The basic equation for the gas-chromatography method is

$$\mathcal{D}_{12} \left[\frac{\partial^2 x}{\partial z^2} + \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial x}{\partial r} \right) \right] - U(r) \frac{\partial x}{\partial z} = \frac{\partial x}{\partial t}, \quad (3.2-13)$$

where x is the mole fraction of the pulse component, U is the velocity of the carrier gas, z is the axial distance, r is the radial coordinate of the tube, R_0 is the constant radius of the tube, and t denotes time. The boundary conditions are:

$$(\partial x / \partial r)_{r=0} = 0,$$

and

$$(\partial x / \partial r)_{r=R_0} = 0.$$

The initial condition depends on the pulse shape at the injection point. As for the point-source method, gas-chromatography involves the assumptions of constant pressure and temperature, constant \mathcal{D}_{12} , one-dimensional flow, and axial symmetry. Additional assumptions are as follows:

(1) The carrier flow velocity is laminar with a parabolic profile; i.e., $U(r) = 2\bar{U}[1 - (r/R_0)^2]$, where \bar{U} is the average velocity.

(2) Convection effects are absent.

(3) The initial pulse of sample may be well approximated by a delta-function.

Subject to these conditions and assumptions, the solution of eq (3.2-13) is given by

$$\bar{x} = \frac{N}{2\pi R_0^2 n} (\pi \mathcal{D}_{\text{eff}} t)^{-1/2} \exp \left[\frac{-(z - \bar{U}t)^2}{4\mathcal{D}_{\text{eff}} t} \right], \quad (3.2-14)$$

where \bar{x} denotes the mean mole fraction of sample in a cross section, n is the total number density, N is the number of molecules of the pulse injected into the carrier gas at $z=0$, $t=0$, and \mathcal{D}_{eff} denotes the effective diffusion coefficient,

$$\mathcal{D}_{\text{eff}} = \mathcal{D}_{12} + R_0^2 \bar{U}^2 / 48 \mathcal{D}_{12}. \quad (3.2-15)$$

The first term on the right-hand side of eq (3.2-15) accounts for the dispersion of the pulse caused by diffusion in the axial direction. The second term is known as the Taylor diffusion coefficient, and accounts for the dispersion of the pulse caused by the parabolic velocity profile, as modified by diffusion in the radial direction.

The experimental procedure is to observe the concentration \bar{x} as a function of time at the end of the tube ($z=L$); eq (3.2-14) for this case is

$$\bar{x}(t) = \frac{N}{2\pi V} \left[\pi \left(\frac{\mathcal{D}_{\text{eff}}}{\bar{U}L} \right) \left(\frac{\bar{U}t}{L} \right) \right]^{-1/2} \exp \left[\frac{-(1 - \bar{U}t/L)^2}{4 \left(\frac{\mathcal{D}_{\text{eff}}}{\bar{U}L} \right) \left(\frac{\bar{U}t}{L} \right)} \right], \quad (3.2-16)$$

where $V = \pi R_0^2 L$ is the tube volume. This is a skewed, not Gaussian, distribution, but if $\mathcal{D}_{\text{eff}} \bar{U}L \leq 0.01$, the distribution becomes nearly Gaussian [34]. The reason is that $\bar{U}t/L$ must be nearly unity when $\mathcal{D}_{\text{eff}} \bar{U}L$ is small, or else $\bar{x}(t)$ becomes too small to measure accurately. In the approximation that $\bar{U}t/L \approx 1$, eq (3.2-16) becomes a Gaussian, with variance τ given by

$$\tau^2 = \frac{2\mathcal{D}_{\text{eff}}}{\bar{U}L} = \frac{2\mathcal{D}_{12}}{\bar{U}L} + \frac{R_0^2 \bar{U}}{24\mathcal{D}_{12}}. \quad (3.2-17)$$

A simple way to determine the variance is to measure the peak width at half height, $w_{1/2}$, related to τ as

$$w_{1/2} = 2(2 \ln 2)^{1/2} \tau. \quad (3.2-18)$$

The calculation of \mathcal{D}_{12} from a measured value of $w_{1/2}$ requires solution of a quadratic equation, which has two roots; one root corresponds to the physical value of \mathcal{D}_{12} when $\bar{U} \leq (48)^{1/2} \mathcal{D}_{12} / R_0$, and the other root when $\bar{U} > (48)^{1/2} \mathcal{D}_{12} / R_0$. An experimental check is that calculated values of the physical \mathcal{D}_{12} must be independent of \bar{U} .

In addition to the normal instrumentation errors for flow, temperature, etc., the gas-chromatography method has an appreciable error contribution from the measurement of the peak width at half height of the dispersion profile. Values of $w_{1/2}$ have been obtained with precisions of about 1 percent, but in terms of \mathcal{D}_{12} this level of precision is degraded because of the quadratic relationship between $w_{1/2}$ and \mathcal{D}_{12} .

Possible uncertainties of gas chromatography are as follows:

(1) Entrance effects caused by the injection of a finite volume of sample into the carrier gas. It is in principle impossible to inject a delta function of sample into the carrier, though in practice the time of injection may be quite short and the sample volume small. Entrance effects can be accounted for by a short correction tube of precisely the same diameter as the regular long-tube [35]. The use of both a long-tube and a short-tube also corrects for effects of stagnant volumes associated with injection and detection devices, and connections along the tube. Instead of two columns, two detectors in one column may be used to eliminate entrance effects [36, 37]. In other studies ingenious sample injection devices have been used. The sample volume should be less than about 1 percent of the tube volume.

(2) Nonsymmetrical dispersion characteristics caused by disturbances to the velocity profile. These disturbances may be due to variations in the carrier gas flow rate or to rough tube surfaces. Some tubes are made in coil form, and bending the tube tends to produce higher velocities on the inside radius than the outside radius. The magnitude of this effect depends on the radius of curvature of the coil. At the detector the concentration curve may be skewed for values of $\mathcal{D}_{\text{eff}}/\bar{U}L \geq 0.01$.

(3) Small pressure drops caused by viscous flow in long tubes and by interferences to flow due to detector(s) immersed in the stream.

(4) End effects caused by the detection of the sample dispersion characteristics. Detector elements may disturb the concentration profile, may not measure point values of the concentration profile, but a finite amount of sample, and may not give a linear response to concentration.

The inaccuracies of determinations of \mathcal{D}_{12} by gas chromatography have been reported to be about 1 to 2 percent. The reliability of these results is best estimated by comparisons with \mathcal{D}_{12} by other methods. At a temperature of 300 K comparisons show deviations up to 4 percent, with an average deviation of about 2 percent. At temperatures up to 500 K the deviations are within 5 percent. Thus, results by gas chromatography are considered to have the same overall level of reliability as the point-source method, that is, uncertainties within 5 percent.

e. Evaporation Tube

In 1873 Stefan developed the evaporation-tube method, which is useful for determinations of \mathcal{D}_{12} for vapor-gas mixtures. The method has been extensively used by other investigators, and until recently these studies have produced almost all the values of \mathcal{D}_{12} for vapor-gas mixtures. The idea of the method is simple. The evaporation rate of a liquid which partially fills a tube is controlled by diffusion through the stagnant gas which fills the rest of the tube. The diffusion coefficient can be determined from observations of the (slow) loss of liquid from the tube at constant temperature and pressure.

In this method the liquid to be volatilized is placed at one end of a vertical tube, the other end of which is open. The tube is a cylinder of uniform cross section, and usually with the approximate dimensions of 5 to 10 mm in diameter and 10 to 20 cm in length. From the gas-liquid interface, vapor diffuses through the gas to the mouth of the tube. At the interface the mixture composition depends on the vapor pressure of the liquid. Across the tube outlet gas flows and carries the vapor away. The rate of liquid loss is observed over long periods of about half a day in order to determine values of \mathcal{D}_{12} .

The same procedure is applicable to the volatilization of a solid in place of a liquid in the evaporation tube.

The evaporation-tube method involves a simple experimental technique, but the studies are restricted to narrow intervals of temperature which are strongly dependent on the volatility of the substance to be tested. The evaporation-tube results for \mathcal{D}_{12} are available for hundreds of different gas pairs. The studies are listed in chronological order in table 7. Of these publications, which comprise

more than seventy articles, about one-third have been published since 1960.

The simple theory for the evaporation-tube method involves the usual assumptions of constant pressure and temperature, constant \mathcal{D}_{12} , one-dimensional diffusion, axial symmetry, and the absence of convection effects. It also involves additional assumptions as follows:

(1) Quasi-steady-state conditions. This assumption means that the composition gradients between the liquid level and the tube outlet are constant. Since the rate of evaporation is slow the gas-vapor column in the tube changes little in height, and even though there are steady losses of liquid due to evaporation the diffusion path can be approximated as constant. A constant liquid level could be maintained, for example, by adding liquid to the evaporation tube at a rate equal to losses due to evaporation. Under quasi-steady-state conditions the flux of vapor is constant.

(2) Gas insolubility. The gas does not dissolve into the liquid. It follows from these assumptions that the gas in the tube is stagnant (zero flux); that is, the net flux in the tube consists only of vapor. The fundamental diffusion equations, eqs (2.1-3) and (2.1-4), then become

$$J_1 = -n\mathcal{D}_{12}(\partial x_1/\partial z) + x_1(J_1 + J_2), \quad (3.2-19)$$

$$J_2 = 0, \quad (3.2-20)$$

where subscript 1 denotes the vapor and subscript 2 the gas. The boundary conditions of the system are that the vapor concentrations are constant at the gas-liquid interface, $(x)_0$, and at the outlet of the tube $(x)_L$. In eq (3.2-19) the vapor velocity profile has been implicitly assumed to be flat, in accordance with the assumption of one-dimensional diffusion. The integration of eq (3.2-19) gives

$$J_1 = (n\mathcal{D}_{12}/L) \ln \left[\frac{1 - (x)_0}{1 - (x)_L} \right], \quad (3.2-21)$$

where the axial distance is measured from the gas-liquid interface, $z=0$, and at the tube outlet $z=L$. The experimental procedure does not require knowledge of the vapor composition as a function of distance, but only the net loss of vapor from the tube. Since the liquid level or the diffusion path length actually changes slowly, the flux of vapor can be related to this change by

$$(dL/dt) = J_1 M / N_0 \rho_{\text{liq}}, \quad (3.2-22)$$

where M is the molecular weight of the liquid, N_0 is Avogadro's number, and ρ_{liq} is the density of the liquid. In order to obtain a final expression useful for the calculation of \mathcal{D}_{12} , the following additional assumptions are made:

(1) The vapor concentration at the gas-liquid interface, $(x)_0$, corresponds to the equilibrium vapor pressure at the liquid surface temperature.

(2) The vapor concentration at the outlet of the tube, $(x)_L$, is zero. This means that the carrier gas (supplied free of vapor) removes all the vapor away from the outlet.

(3) The gases and vapors are ideal, so that compositions may be expressed in terms of partial pressures.

TABLE 7. Determinations of \mathcal{D}_{12} by the evaporation-tube method^a

Author(s)	Date	Author(s)	Date
Stefan.....	1873	Rossié.....	1953
Baumgartner.....	1877 a,b	Lee and Wilke.....	1954
Guglielmo.....	1881, '82	Bose and Chakraborty.....	1955-56
Winkelmann.....	1884 a,b,c	Carmichael et al.....	1955 a, b
	'85, '88, '89	Cummins and Ubbelohde.....	1955
Stefan.....	1889, '90	Cummings et al.....	1955
Griboiedov.....	1893	Narsimhan.....	1955
Houdaille.....	1896	Raw.....	1955
Naccari.....	1909	Crider.....	1956
Mache.....	1910	Call.....	1957
Naccari.....	1910	Clarke and Ubbelohde.....	1957
Vaillant.....	1911	Richardson.....	1959
Pochettino.....	1914	Altshuler and Cohen.....	1960
Gaede.....	1915	Hudson et al.....	1960
LeBlanc and Wuppermann.....	1916	Jorgensen and Watts.....	1961
Mack.....	1925	Reamer and Sage.....	1963
Topley and Whytlaw-Gray.....	1927	Grieverson and Turkdogan.....	1964
Summerhays.....	1930	Heinzelmann et al.....	1965
Trautz and Ludwig.....	1930	Kohn and Romero.....	1965
Trautz and Ries.....	1931	Stevenson.....	1965
Ackermann.....	1934	Brockett.....	1966
Gilliland.....	1934	Mehta.....	1966
Trautz and Müller.....	1935	Ben-Aim et al.....	1967
Chambers and Sherwood.....	1937	Byrne et al.....	1967
Schirmer.....	1938	Galloway and Sage.....	1967
Brookfield et al.....	1947	Getzinger and Wilke.....	1967
Klotz and Miller.....	1947	Krol et al.....	1967
Goryunova and Kuvshinskii.....	1948	Mikhailov and Kochegarova.....	1967
Gush.....	1948	Nafikov and Usmanov.....	1967
McMurtie and Keyes.....	1948	Pryde and Pryde.....	1967
Hippenmeyer.....	1949	Yuan and Cheng.....	1967
Schwartz and Brow.....	1951	Khomchenkov et al.....	1968
Cvetanović and LeRoy.....	1952	Lugg.....	1968
Kimpton and Wall.....	1952	Mrazek et al.....	1968
Schlinger et al.....	1952-53	O'Connell et al.....	1968
Cummings and Ubbelohde.....	1953	Spencer et al.....	1969

^a Complete reference information is given in Bibliography I.

Under these assumptions, and when eq (3.2-21) is substituted in eq (3.2-22) and integrated, the expression for \mathcal{D}_{12} is

$$\mathcal{D}_{12} = \frac{(L_2^2 - L_1^2)}{2(t_2 - t_1)} \left(\frac{RT}{p} \right) \left(\frac{\rho_{\text{liq}}}{M} \right) \ln \left(\frac{p - p_s}{p} \right), \quad (3.2-23)$$

where p is the total pressure, p_s is the vapor pressure, R is the ideal gas constant, and subscripts 1 and 2 on L and t denote the initial and final times for the observations of the evaporation losses during an interval of diffusion. In some studies the weight loss of liquid is measured instead of the change in height.

The quasi-steady-state condition will be approached within 1 percent for diffusion times greater than $L^2/2\mathcal{D}_{12}$; to satisfy this condition experiments are usually run for several hours [39].

The calculated values of \mathcal{D}_{12} should be corrected for end effects caused by surface tension at the gas-liquid interface and turbulence at the tube outlet. End effects are related to the accurate determinations of the length of the diffusion path. The principal factor is turbulence which arises from interference by the end of the tube to the carrier gas flow. To avoid the effects of turbulence the gas flow rate can be empirically adjusted—not too great to cause large eddy currents, and not too small to cause a nonzero vapor concentration at the tube outlet. The presence of eddy currents will effectively shorten the diffusion path length. An end correction can be made by a graphical

procedure. The calculated values of \mathcal{D}_{12} are plotted as a function of the reciprocal of the observed length of the diffusion path L ; the corrected values of \mathcal{D}_{12} are taken at the extrapolated point, $1/L=0$.

In addition to the normal instrumentation errors, the possible uncertainties of the evaporation-tube method are as follows. The evaluation of the term $\ln[(p-p_s)/p]$ indicates that small changes in pressure and temperature will cause large uncertainties in \mathcal{D}_{12} [38]. Significant variations in barometric pressure and in system temperature may occur since evaporation-tube experiments usually run for many hours. For example, if the partial pressure p_s is 25 torr then for variations of ± 10 torr in total pressure the variation of $\ln[(p-p_s)/p]$ is 1.4 percent for total pressures at about 1 atm. The variations of the liquid surface temperatures may be even more critical because of the sensitivity of the vapor pressure to small temperature changes. For precise results the variation in temperature of the liquid should be no greater than ± 0.1 K.

Other possible uncertainties for the evaporation-tube method are as follows:

(1) Convection effects caused by the direction of vapor diffusion [39]. For example, water-air values of \mathcal{D}_{12} may differ by about 2 percent depending on whether the water is placed in the bottom or at the top of the tube. Additional convection effects are dependent on the diameter of the evaporation-tube, and a possible error of 4 percent is indicated if the diameter of the tube is large.

(2) Nonequilibrium conditions may exist because of excessive rates of evaporation and supercooling at the surface of the liquid.

(3) The contamination of the liquid by trace amounts of impurities may significantly effect the evaporation rate. In a study of water-air, for example, any traces of oil in the air could accumulate on the surface of the water. Another contamination process is due to gases dissolved in the liquid. In one case this effect caused differences of about 5 percent in values of \mathcal{D}_{12} [40]. This shows that liquids should be degassed prior to their use.

(4) The equilibrium values used in the formulas for \mathcal{D}_{12} may themselves have significant uncertainties, especially equilibrium values that have been found in handbooks where the reliability of the data has not been specified.

(5) Many vapors are nonideal gases, and any deviations from ideal-gas behavior affect the accurate specification of the mixture composition [41].

(6) The assumption of a flat velocity profile is not strictly correct, since a parabolic profile develops as the vapor moves away from the gas-liquid interface. However, the maximum effect is only 1.4 percent in deviations of the radial concentration from a uniform (flat) value [42, 43].

At best, the reliabilities of \mathcal{D}_{12} by the evaporation-tube method are several percent. The following cases are illustrative. In thirteen independent studies for the system water-air, the standard deviation is 7.5 percent at 298 K, and at higher tempera-

tures the scatter in the data is even greater [44]. In another review of \mathcal{D}_{12} for water-air, four of twelve studies were dropped from the calculation of the average value because they seemed obviously in error [39]. There are only a few other gas pairs, $\text{H}_2\text{-H}_2\text{O}$ and benzene-air, which have several independent measurements useful for the estimation of reliability. These results are also disappointing.

3.3. Minor Experimental Methods

In this section the six minor methods listed in table 2 are briefly discussed. The minor methods could probably yield more reliable values of \mathcal{D}_{12} by further developments of each. The studies using these methods are listed in table 8, and this listing contains the reference information for this section unless special footnotes are given in the text.

a. Open Tube

If the top of a gas container is opened, the gas will diffuse into the surrounding atmosphere, and analysis of the composition of the remaining gas after a known time permits the determination of \mathcal{D}_{12} . In 1882 the first such measurements were independently made by von Obermayer and by Waitz. The theory of the experiment was described by Stefan in 1871 [45]. Detailed analyses of the method and these early studies have been published [46, 47]. The open-tube method has recently been revived and improved by Frost.

TABLE 8. Determinations of \mathcal{D}_{12} by Minor Experimental Methods^a

Author(s)	Date	Author(s)	Date
a. Open Tube		Grob and El-Wakil.....	1969
von Obermayer.....	1882 a	e. Diffusion Bridge	
Waitz.....	1882 a, b	Buckingham.....	1904
von Obermayer.....	1883, '87	Wicke and Kallenbach.....	1941
Toepler.....	1896	Weisz.....	1957
Foch.....	1913	Bendt ^b	1958
Barus.....	1924 a, b	Scott and Cox.....	1960
Currie.....	1960	Evans et al.....	1961
Frost.....	1967	Wicke and Hugo.....	1961
Kaufmann.....	1967	Scott and Dullien.....	1962
Rhodes and Amick.....	1967	Evans et al.....	1962, '63
Zhukhovitskii et al.....	1968	Kosov and Kurlapov ^b	1966
b. Back Diffusion		Coates and Mian.....	1967
Harteck and Schmidt.....	1933	Henry et al.....	1967
Van der Held and Miesowicz.....	1937	Mian.....	1967
Spier.....	1939, '40	Reist.....	1967
c. Capillary Leak		Zhargasov and Kosov ^b	1968
Klibanova et al.....	1942	Ellis and Holsen.....	1969
Kosov.....	1957	Mian et al.....	1969
Vyshenskaya and Kosov.....	1959, '65	Hawtin et al.....	1969
Kosov and Karpushin.....	1966	Schneider and Schäfer.....	1969
De Paz et al.....	1967	Kosov and Zhargasov ^b	1970
d. Unsteady Evaporation		f. Dissociated Gases	
Mullaly and Jacques.....	1924	Krongelb and Strandberg.....	1959
Mackenzie and Melville.....	1932, '33	Wise.....	1959, '61
Arnold.....	1944	Walker.....	1961
Fairbanks and Wilke.....	1950	Young.....	1961
Nelson.....	1956	Morgan and Schiff.....	1964
de Nordwall and Flowers.....	1958	Yolles and Wise.....	1968
Nikolaev and Aleskovskii.....	1964	Khouw et al.....	1969
Petit.....	1965	Sancier and Wise.....	1969
		Baker.....	1970 b
		Yolles et al.....	1970

^a Complete reference information is given in Bibliography I.

^b The investigations by Bendt involved an apparatus with one capillary, and those by Kosov et al. a bundle of capillaries; all other diffusion-bridge apparatuses used porous septa.

b. Back Diffusion

Harteck and Schmidt in 1933 performed the first low-temperature determinations of \mathcal{D}_{12} , down to 20 K, for a mixture of para-hydrogen in normal hydrogen. The method is an ingenious steady-state flow technique in which one component diffuses upstream against the second flowing component. The composition at one or more upstream points can be used to determine \mathcal{D}_{12} . Back diffusion can be used for diffusion measurements at extreme temperatures, low or high, as well as for "tagged" molecules. The description of the original method has been translated, in part, into English [2].

c. Capillary Leak

The capillary-leak method is suitable for measurements of \mathcal{D}_{12} over a large range of temperatures because it involves no moving parts. In 1942 this method was first used by Klibanova et al. to determine \mathcal{D}_{12} at high temperatures, up to 1533 K. In 1967 De Paz et al. determined the self-diffusion coefficient of Ar at low temperatures, down to 78 K. Except for the results by De Paz et al., the precision and reliability of \mathcal{D}_{12} obtained by the capillary-leak method have been poor.

d. Unsteady Evaporation

An alternate evaporation-tube method was developed by Arnold in 1944. His purpose was to obtain a quantitative basis for calculations of unsteady-state vaporization of a liquid into a gas, a process of industrial importance. The equations obtained also furnished a basis for relatively quick determinations of \mathcal{D}_{12} for vapor-gas mixtures. Measurements could be made in minutes, not in hours as required by the Stefan evaporation tube. The reliability of the unsteady-evaporation method is probably slightly better than for the evaporation tube, but more meaningful comparisons are not possible because of the meager data available. A somewhat similar technique was used earlier by Mackenzie and Melville with bromine vapor. Other unsteady-evaporation studies are also listed in table 8.

e. Diffusion Bridge

This is a steady-state flow method in which two gas streams flow across opposite ends of a hollow capillary tube or opposite faces of a porous septum, and the emerging streams are analyzed. The flow rates are controlled, and adjusted to produce any desired pressure difference across the capillary. The ends of the capillary are generally maintained at equal total pressures, thus in the capillary, or septum, there is uniform pressure and no viscous flow. The advantage of the diffusion bridge is that no valves are required in the zone of the apparatus where diffusion occurs, so that the method is amenable to operation over wide temperature ranges.

The diffusion bridge has been used only once with a capillary, to obtain absolute values of \mathcal{D}_{12} down to 1.74 K. This work was done by Bendt in 1958.

The diffusion bridge has been used frequently with a porous septum, to obtain relative values of \mathcal{D}_{12} up to 882 K. These studies require the calibration of the porosity of the system by means of independently published values of \mathcal{D}_{12} .

f. Dissociated Gases

Direct measurements of the diffusion of highly reactive species such as free radicals and valence-unsaturated atoms are difficult, but are needed for basic understanding of many phenomena in chemical reactions and at high temperatures. There are a variety of techniques, both of the flow and non-flow types, that have been used to measure the diffusion of H, N, and O atoms in different gases. The results for \mathcal{D}_{12} extend over a temperature range of 202 to 873 K. Dissociated gases were first studied in 1959 by Wise and by Krongelb and Strandberg. The technique by Morgan and Schiff minimizes uncertainties due to chemical reactions; the complete neglect of reactions in calculations of \mathcal{D}_{12} introduces less than 1 percent error. The results for dissociated gases, as might be expected, are not very reproducible; the results show a scatter of about 10 percent or more for many gas pairs.

In some cases there are indirect methods available for the determination of \mathcal{D}_{12} for dissociated gases which probably give more reliable results than the present direct methods. For example, \mathcal{D}_{12} for H-H₂ can be obtained from measurements of the mixture viscosity [48], and \mathcal{D}_{12} for N-N₂ and O-O₂ at $T > 1000$ K can be obtained from molecular-beam scattering experiments and semi-empirical quantal calculations [49].

3.4. Miscellaneous Experimental Methods

This section briefly describes several miscellaneous methods that have been used to determine values of \mathcal{D}_{12} . A list of these studies is given in table 9, and this listing contains the reference information for this section unless special footnotes are given in the text. The listing is not comprehensive, as a complete enumeration of all miscellaneous methods used at some time or another would be both futile and boring. The miscellaneous methods listed have both general applicability and experimental ingenuity.

a. Droplet Evaporation

Observations of the rate of evaporation of a small sphere of volatile material may be utilized to determine vapor-gas \mathcal{D}_{12} . The applicable theory is similar to the evaporation tube. Droplet evaporation studies have been made for water, for heavy organic chemicals, and for iodine in air.

b. Dufour Effect

When different gases mix, a small temperature gradient is set up; this is called the Dufour effect or the diffusion thermoeffect. The asymptotic time decay of the temperature gradient can be used to determine \mathcal{D}_{12} , and results are available for about ten gas pairs at 293 K. These results have varied agreements, within 10 percent, with \mathcal{D}_{12} obtained by the major methods.

c. Thermal Separation Rate

The inverse of the Dufour effect is thermal diffusion, in which an imposed temperature gradient causes the components of a mixture to separate. The rate at which an initially uniform mixture separates under an imposed temperature gradient can be used to determine \mathcal{D}_{12} . The results, however,

TABLE 9. Determinations of \mathcal{D}_{12} by miscellaneous experimental methods^a

Author(s)	Date	Author(s)	Date
a. Droplet Evaporation		f. Cataphoresis	
Langmuir.....	1918	Freudenthal.....	1966
Topley and Whytlaw-Gray.....	1927	Hogervorst and Freudenthal.....	1967
Houghton.....	1933	Hogervorst.....	1971
Bradley et al.....	1946	g. Resonance Methods	
Bradley and Shellard.....	1949	(Nuclear Magnetic Resonance)	
Birks and Bradley.....	1949	Luszczynski et al.....	1962
Bradley.....	1951	Lipsicas.....	1962
Bradley and Waghorn.....	1951	Hartland and Lipsicas.....	1963
Katan ^b	1969	Luszczynski et al.....	1967
b. Dufour Effect		Khoury and Kobayashi.....	1970
Waldmann.....	1944, '47	(Optical Pumping)^c	
Mason et al.....	1967	Franzen.....	1959
c. Thermal Separation Rate		Bernheim.....	1962
Nettley.....	1954	McNeal.....	1962
van Itterbeek and Nihoul.....	1957	Anderson and Ramsey.....	1963
Lonsdale and Mason.....	1957	Legowski.....	1964
Saxena and Mason.....	1959	Ramsey and Anderson.....	1964
Weissman et al.....	1961	Bernheim and Korte.....	1965
Wendt et al.....	1963	Gozzini et al.....	1967
Mason et al.....	1964 b	(Mercury Band Fluorescence)	
d. Kirkendall Effect		Coulliette.....	1928
McCarty and Mason.....	1960	Biondi.....	1953
Mason.....	1961	McCoubrey.....	1954
e. Sound Absorption		McCoubrey and Matland.....	1954
Holmes and Tempest.....	1960	Matland and McCoubrey.....	1955
Carey et al.....	1966	McCoubrey and Matland.....	1956
Carey et al.....	1968	Tubbs.....	1967

^a Complete reference information is given in Bibliography I.

^b This is a clever combination of a uniform-pressure experiment with a porous membrane and the evaporation-tube method; the motion of an evaporating liquid head in a tube is used to determine the diffusion coefficient.

^c A recent review article has been published by Violino (1968).

are not very reliable. The principal uncertainties arise from averaging the temperatures of the imposed temperature gradient and geometrical factors of the apparatus.

d. Kirkendall Effect

In solids, the net drift of inert markers placed near a diffusion interface is called the Kirkendall effect. A similar effect exists in gases, and the speed of the marker motion can be used to determine \mathcal{D}_{12} . The marker is located in a tube connected in parallel to a two-bulb apparatus. A value of \mathcal{D}_{12} has been obtained for He-Ar at 303 K, which is in excellent agreement with directly determined \mathcal{D}_{12} .

Smoke particles suspended in a diffusing gas mixture can also be used as Kirkendall markers [50, 51].

The diffusion pressure-effect, discussed in section 2.1, part a, is closely related to the Kirkendall effect. It could therefore also be used to determine values of \mathcal{D}_{12} , but this has not yet been done.

e. Sound Absorption

The passage of a sound wave through a gas mixture produces a local partial separation of the components, caused mostly by pressure diffusion. The remixing by diffusion is out of phase with the sound wave, and the absorption of an ultrasonic wave in a gas mixture is stronger than in either pure component. The excess absorption depends on \mathcal{D}_{12} , which in principle can then be determined [52].

This method has been tested on the gas pair He-Ar up to temperatures of ~ 5000 K. The agreement of these results with other available data is good at 300 K, but poor between 1255 and 4990 K. These measurements are difficult to perform and the results at high temperatures are scattered.

f. Cataphoresis

A dc-discharge in a gas mixture causes a partial separation of components. The phenomenon, which also occurs in solutions, is called cataphoresis. The separation disappears by diffusion after the discharge is stopped, and \mathcal{D}_{12} can be calculated from the rate of disappearance of the separation. At the time of this evaluation cataphoresis had been used only for the gas pairs Ne-Ar from 300 to 650 K; the results are in good agreement with other direct measurements. While this work was in the process of publication further results were published for all the noble gas pairs, except Kr-Xe, from 300 to 1400 K [53]; but these results were not evaluated in this report.

g. Resonance Methods

The principle of all resonance methods is to "tag" some of the molecules in a gas, and then follow their dispersion due to diffusion. The tags used have been such things as the orientation of nuclear spin (nuclear magnetic resonance), the population of magnetic sublevels in the ground state (optical pumping), or a metastable excited

electronic state (mercury band fluorescence). The names in parentheses indicate the groupings for the studies listed in table 9. The nuclear magnetic resonance technique has been used to determine \mathcal{D}_{12} at very low temperatures, down to 20 K for mixtures of ortho- and para-hydrogen, and down to 1.13 K for the self-diffusion coefficient of ^3He . The optical pumping technique has been used to determine \mathcal{D}_{12} of alkali metal vapors (Na, Rb, and Cs) in various other gases.

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4. Treatment of Data

In this section are outlined the procedures used to evaluate the entire body of experimental data, and the derivation of semi-empirical approximations for the composition dependence and temperature dependence of \mathcal{D}_{12} .

The original data, published over the last one hundred years, were compiled by author and by gas pair. Bibliography I lists the references to experimental studies by author, and the gas pairs investigated and the experimental methods are noted. A cross-listing of \mathcal{D}_{12} by gas pair is given in table 16, in which the temperature range is also noted. In addition, there are bibliographies for short-range and long-range interaction data, and for measured mixture viscosities.

4.1. Reliability Estimates

The critical evaluation of the reliability of \mathcal{D}_{12} from direct measurements included the following factors:

- (1) experimental method,
- (2) reproducibility of \mathcal{D}_{12} by different experimental methods or laboratories,
- (3) precision and number of measurements from a given laboratory, and
- (4) temperature dependence measured.

For indirect measurements, the reliability of \mathcal{D}_{12} considered the reported accuracy of other transport property data of mixtures—viscosity, thermal conductivity, and thermal diffusion factor—and of molecular beam measurements. Whenever collision integral ratios were employed in intermediate calculations of \mathcal{D}_{12} , their reported accuracy was considered as well as the choice of the potential model.

For all measurements, the results of the more recent studies were *not* assumed to be necessarily more accurate than those from earlier studies. All the data for each gas pair were carefully inspected for discrepancies and systematic errors with the aid of large-scale graphs. From these it became ap-

parent that the small composition dependence of \mathcal{D}_{12} had to be taken into account, and that compact means of summarizing values of \mathcal{D}_{12} over large temperature ranges were necessary.

The assignment of reliability estimates to experimental data always involves a large measure of subjective judgment [1-3].⁴ Even after consideration of such things as reproducibility and internal consistency, external consistency for different types of apparatus and for different workers in different laboratories, and so on, the final decisions are nevertheless based heavily on the judgments of the evaluators. An attempt has been made to be conservative, in order that there shall be a high probability that the "true" value of a diffusion coefficient lies within the specified range of uncertainty. An attempt has also been made to be fair and not arbitrarily downgrade good measurements, but it is quite possible that a particular \mathcal{D}_{12} may be more accurate than is implied by the specified uncertainty limits which are given in section 5.

4.2. Correction for Composition Dependence

Even though the magnitude of the composition dependence of \mathcal{D}_{12} is relatively small, from 0 to 5 percent for all gas pairs considered, the effect is sometimes greater than the uncertainty of experimental measurements. The formulas for the composition dependence according to the second approximation of the Chapman-Enskog theory (sec. 2.4) are cumbersome to use, especially when thousands of data points must be considered. A simpler and more convenient formula of sufficient accuracy can be developed as follows.

The entire composition dependence of \mathcal{D}_{12} is contained in the small term Δ_{12} , given in section 2.4, which depends on both temperature and composition. The major complication of the Chapman-Enskog expression for Δ_{12} is its composition-dependent part (containing the P 's and Q 's). Previous work [4] indicates that the composition dependence can be adequately approximated by the formula,

$$\Delta_{12} \approx \zeta (6C_{12}^* - 5)^2 \frac{ax_1}{1 + bx_1}, \quad (4.2-1)$$

where ζ is a numerical constant between 1 and 2, x_1 is the mole fraction of the heavy component, and a and b are

$$a = \frac{1}{10} (-S_2/Q_2) (P_1/S_1), \quad (4.2-2)$$

$$b = (-S_2/Q_2) (Q_1/S_1) - 1. \quad (4.2-3)$$

The S_1 and S_2 occur in the expression for the thermal diffusion factor (see sec. 2.7), which is related theoretically to Δ_{12} [4]. This formula is most accurate for $M_1 \gg M_2$; the lower accuracy for $M_1 \approx M_2$ is not important, however, because Δ_{12} is negligible in such cases.

The expressions for a and b can be further simplified. It is obvious that

$$b + 1 = 10a(Q_1/P_1). \quad (4.2-4)$$

Substitution for P_1 and Q_1 from eqs (2.4-1) and (2.4-3) yields

$$Q_1/P_1 = \left(\frac{5}{2} - \frac{6}{5} B_{12}^*\right) + 3(M_2/M_1)^2 + \frac{8}{5} A_{12}^* (M_2/M_1). \quad (4.2-5)$$

An adequate approximation for the present purpose is to take $B_{12}^* = 5/4$ (Kihara approximation) and $A_{12}^* \approx 1.1$, which yields

$$b + 1 = 10a(1 + 1.8m + 3m^2), \quad (4.2-6)$$

where

$$m = M_2/M_1 < 1. \quad (4.2-7)$$

For small m , the major variation of a comes from the factor $(-S_2/Q_2)$ and can be represented by the simple expression

$$a \approx \frac{2^{1/2}}{8(1 + 1.8m)^2} \frac{\bar{\Omega}_{12}^{(1,1)}}{\bar{\Omega}_{22}^{(2,2)}}. \quad (4.2-8)$$

The factor $(1 + 1.8m)^2$ is an empirical representation of the various mass dependences, but the rest of the expression comes from theory. The collision integrals in a may be obtained either by calculation from a potential model or from experimental values of \mathcal{D}_{12} and η_2 , the viscosity of the light component, whereby eq (4.2-8) may be written as

$$a \approx \frac{3(1+m)^{1/2}}{20(1+1.8m)^2} \frac{\eta_2 RT}{p \mathcal{D}_{12} M_2}. \quad (4.2-9)$$

The quantities a and b vary only weakly with temperature, and can usually be taken as constant.

The complete result for Δ_{12} is thus given by eqs (4.2-1), (4.2-6), (4.2-7), and (4.2-8) or (4.2-9). The value of C_{12}^* in eq (4.2-1) can be calculated from the Lennard-Jones (12-6) potential, and depends only on the temperature and the value of ϵ ; the results are not too sensitive to the choice of the potential and the precise value of ϵ used. The value of ζ is unity according to the Chapman-Enskog second approximation; since this approximation seems to underestimate Δ_{12} , it is better to take ζ from experiment if accurate data are available. Values of ζ , a , b , and ϵ are listed in table 15 of section 5.

Figure 3 shows a comparison of values of Δ_{12} calculated from the semi-empirical approximation with values calculated from the Chapman-Enskog expression. Two mass ratios are shown, which represent reasonable values for ordinary gas pairs like He-Ar and Ne-Ar; the potential parameters used in the calculations correspond to these two gas pairs. A high reduced temperature of $kT/\epsilon = 10$ is used, for which Δ_{12} is large. The results for Δ_{12} are in agreement within the uncertainties of experimental measurements.

Values of the empirical constant ζ have been determined from measurements of the composition dependence of \mathcal{D}_{12} for only fourteen systems (counting H_2 and D_2 as the same); namely, the ten noble gas pairs [5], H_2-N_2 [6], H_2-Ar and H_2-CO_2 [4], and He- N_2 [7]. These are the only systems for which enough accurate data on composition dependence exist to justify assigning ζ a value other than 1.0. As can be seen from table 15 of section 5,

⁴ Figures in brackets indicate the literature references at the end of Section 4.

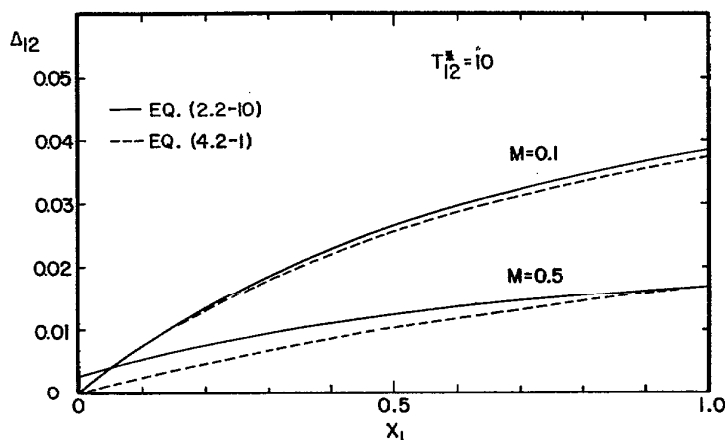


FIGURE 3. Composition dependence of Δ_{12} at two mass ratios (m).

Equation (2.2-10) is the exact formula of the Chapman-Enskog theory; Eq. (4.2-1) is the semi-empirical expression of this report.

the empirically determined values of ζ do lie between 1 and 2, as expected. An advantage of the semi-empirical formula is that improved experimental information on the composition dependence of \mathcal{D}_{12} can be easily accommodated by adjustment of values of ζ .

Experimental \mathcal{D}_{12} data were adjusted to refer to an equimolar composition according to the relation

$$\mathcal{D}_{12}(x_1 = 1/2) = \mathcal{D}_{12}(x_1) \left[\frac{1 + \Delta_{12}(x_1 = 1/2)}{1 + \Delta_{12}(x_1)} \right], \quad (4.2-10)$$

where $\mathcal{D}_{12}(x_1)$ was the value measured at mole fraction x_1 , and the Δ_{12} were calculated from eq (4.2-1) with the constants given in table 15 of section 5. An equimolar basis is a reasonable compromise between composition extremes, and required a minimum number of adjustments of the data.

4.3. Correlation for Temperature Dependence

The temperature dependence of \mathcal{D}_{12} can be correlated by a semi-empirical equation which is applicable over a wide range of temperature. The theoretical background for the correlation, in terms of intermolecular forces, has been presented in section 2.3; the equation itself is an empirical composite of terms corresponding to various types of contributions to the intermolecular forces. The equation correlates the temperature dependence of \mathcal{D}_{12} within the experimental uncertainties of the experimental results with at most four adjustable parameters, and can be put into simplified form for data with low reliability or with limited temperature range.

The background is briefly as follows. Many previous correlations of the temperature dependence of \mathcal{D}_{12} have been published [8-35]. These correlations have usually been restricted to fairly narrow temperature ranges between about 200 and 500 K, because of the lack of data at low and high temperatures. But recent results on \mathcal{D}_{12} by direct measurements and by calculations from molecular-beam scattering experiments have significantly extended

the temperature range, which now extends roughly from about 10 000 down to 80 K, or lower.

In figure 1 the inset shows the characteristic temperature dependence of $p\mathcal{D}_{12}$. Over a narrow range of temperatures a plot of $\ln(p\mathcal{D}_{12})$ versus $\ln T$ is essentially linear, as would be expected for an inverse-power potential, but over a large temperature range such a plot shows curvature. At low temperatures the curvature is caused by the increasing influence of the long-range attractive potential. At high temperatures the curvature is caused by the increasing "softness" of the repulsive potential at small separation distances, as would be expected for an exponential potential.

The foregoing features can be fitted by an equation of the form,

$$\ln(p\mathcal{D}_{12}) = \ln A + s \ln T - \ln [\ln(\varphi_0/kT)]^2 - (S/T) - (S'/T^2), \quad (4.3-1)$$

where A , s , φ_0 , S , and S' are empirical constants, and k is the Boltzmann constant. The double logarithm term is taken from eq (2.3-4) and represents an exponential repulsion potential. The value of φ_0 is taken from independent molecular-beam experiments [36], and is not adjustable; however, its precise value is not critical for correlation purposes, because errors in φ_0 are compensated for by values of s . The values of s are equal to or slightly greater than 3/2, as expected from theory. The terms containing S and S' are Sutherland-Reinganum terms, as in eq (2.3-11), and account for the attractive potential. For most gas pairs S' is not needed and can be taken as zero.

In many cases the values of \mathcal{D}_{12} are not sufficiently precise to require the use of the double logarithm term in eq (4.3-1), and an adequate representation is given by

$$\ln(p\mathcal{D}_{12}) = \ln A + s \ln T - (S/T). \quad (4.3-2)$$

All the data could be correlated within the range of estimated experimental uncertainties by combinations of eqs (4.3-1) and (4.3-2).

An advantage of eqs (4.3-1) and (4.3-2) is that they are linear in all the adjustable constants (A , s , S , and S'), so that least-squares calculations are easy. A disadvantage is that the Sutherland-

Reinganum terms for the effect of the long-range attraction do not permit the equations to be used at very low temperatures, where the London dispersion energy dominates. At present, no measurements of \mathcal{D}_{12} seem to fall in this range, however. The equations are usable only for $kT/\epsilon > 1$, and should never be extrapolated to low temperatures. At very low temperatures \mathcal{D}_{12} has the asymptotic (classical) form,

$$p\mathcal{D}_{12} = AT^{1/6}, \quad T \rightarrow 0, \quad (4.3-3)$$

where A is easily calculated if the London constant C is known [37]. The reliability of eq (4.3-3) has been discussed in section 2.6, part a.

The values of the constants for eqs (4.3-1), (4.3-2), and (4.3-3) are given in tables 12, 13, and 14, respectively, of section 5. Details on their determination are presented in section 5.

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5. Results

In this section the recommended values of \mathcal{D}_{12} are presented. The reliability estimates of these \mathcal{D}_{12} are given in section 5.1. Then, in section 5.2, the constants are listed for the correlations of \mathcal{D}_{12} as a function of temperature and of composition. In section 5.3 the deviations between data and the recommended values of \mathcal{D}_{12} are illustrated by graphs. The detailed remarks on the critical evaluation of data for weighted least-squares calculations are given in section 5.4.

This chapter summarizes the most reliable experimental results for binary gaseous diffusion coefficients through 1968. Gas pairs for which only limited or uncertain data exist are not included here, but these experimental measurements may be retrieved with the aid of table 16, which extends through 1970. (See table 16 at the end of section 5.)

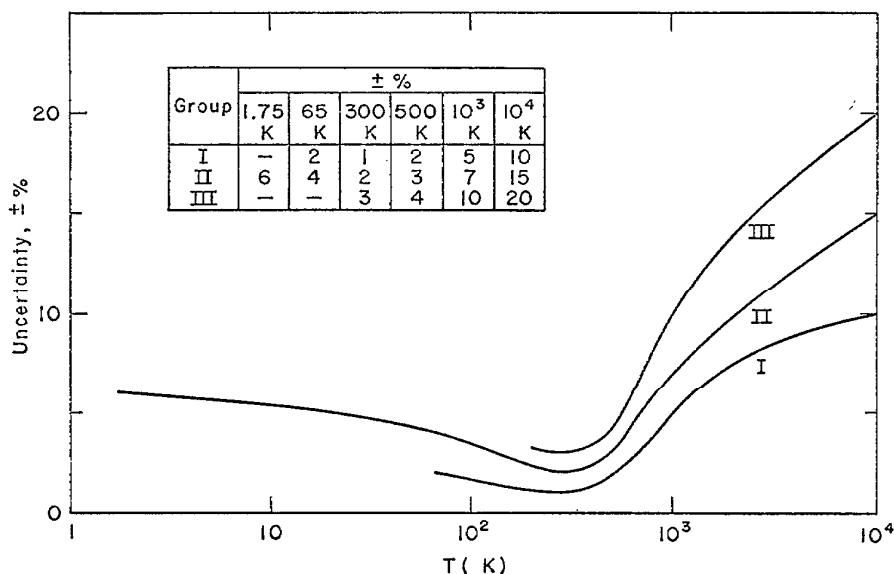
5.1. Uncertainty Limits

The sources of reliable values of \mathcal{D}_{12} are roughly as follows. For all gas pairs the most accurate results are at approximately 300 K, because of the existence of a large number of independent measurements by the most reliable experimental methods. Both closed-tube and two-bulb measurements are usually available at temperatures from 200 to 500 K, and several additional two-bulb measurements exist at lower temperatures. The temperature limits of \mathcal{D}_{12} from direct experiment have been extended in both directions by the use of data on mix-

ture viscosities, or, in a few instances, on thermal diffusion factors. These derived values of \mathcal{D}_{12} have slightly less reliability than those near room temperature. The magnitude of the extended temperature range is variable, and depends on the particular gas pair. For several gas pairs, data are available to about 1000 K, and in a few instances to higher temperatures, but less than 2000 K, from the point-source method. Values of \mathcal{D}_{12} from 1000 to 10 000 K are derived mainly from molecular-beam measurements, which generally have the least reliability.

The gas pairs for which recommended data are given can be grouped into three categories of reliability, as shown in figure 4. A gas pair in Group I, for instance, has uncertainty limits of ± 1 percent in \mathcal{D}_{12} at 300 K; the uncertainty increases to ± 5 percent at 1000 K, and to ± 10 percent at 10 000 K. The temperature dependences of the uncertainty limits are shown in figure 4, and the gas pairs assigned to each group are listed in table 10. The borderline systems are assigned to the higher group, but are noted by a question mark. Table 10 lists the gas pairs in terms of one common member in a series of gas pairs; this leads to some duplication but is helpful for quick reference.

A miscellaneous group of gas pairs is also included because of possible special interest, and their uncertainty limits are listed in table 11. The miscellaneous group contains mixtures with one component water, carbon dioxide, or dissociated gas (H, N, or O).

FIGURE 4. Estimated uncertainty limits of \mathcal{D}_{12} as a function of temperature.TABLE 10. Grouping of systems according to uncertainty limits of \mathcal{D}_{12} .

Group I	He-(Ne,Ar,Kr,Xe) Ne-(He,Ar,Kr,Xe) Ar-(He,Ne,Kr,Xe) Kr-(He,Ne,Ar,Xe?) H ₂ -N ₂ ?
Group II	³ He- ⁴ He He-(H ₂ ,N ₂ ,CO,O ₂ ,air,CO ₂) H ₂ -(He,Ne?,Ar,Kr?,D ₂ ,CO,air,CO ₂) N ₂ -(Ar?,CO,CO ₂)
Group III	Ar-(CH ₄ ,CO,O ₂ ,air,CO ₂ ,SF ₆) H ₂ -(Xe,CH ₄ ,O ₂ ,SF ₆) CH ₄ -(He,Ar,H ₂ ,N ₂ ,O ₂ ,air,SF ₆) N ₂ -(Ne,Kr,Xe,CH ₄ ,O ₂ ,SF ₆) CO-(Ar,Kr,O ₂ ,air,CO ₂ ,SF ₆) O ₂ -(Ar,H ₂ ,CH ₄ ,N ₂ ,CO,CO ₂ ,SF ₆) CO ₂ -(Ar,CO,O ₂ ,air,N ₂ O,SF ₆) SF ₆ -(He,Ar,H ₂ ,CH ₄ ,N ₂ ,CO,O ₂ ,air,CO ₂)
Miscellaneous Systems	H ₂ O-(N ₂ ,O ₂ ,air,CO ₂) CO ₂ -(Ne,H ₂ O,C ₃ H ₈) H-(He,Ar,H ₂) N-N ₂ O-(He,Ar,N ₂ ,O ₂)

TABLE 11. Uncertainty limits for systems of the miscellaneous group

System	T Range, K	Uncertainty, ± %
H ₂ O-N ₂	282 to 373	4
H ₂ O-O ₂	282 to 1070	7
H ₂ O-air	282 to 1070	5 to 10
H ₂ O-CO ₂	296 to 1640	10 to 7
CO ₂ -Ne	195 to 625	3 to 5
CO ₂ -C ₃ H ₈	298 to 550	3 to 5
H-H ₂	~ 300	5
	> 1000	30
H-N ₂ ,O-N ₂ ,O-O ₂	~ 300	10
	> 1000	25
H-He,H-Ar, O-He,O-Ar	~ 300	15
	> 1000	30

The gas pairs of Group I have the most reliable values of \mathcal{D}_{12} for two principal reasons. First, results below 400 K are based on the very careful measurements of van Heijningen et al. Second, at high temperatures, the values of \mathcal{D}_{12} derived from molecular-beam scattering experiments for the noble gas pairs are more reliable than for diatomic or polyatomic gases, as discussed in section 2.6, part b. In Group I the gas pairs Kr-Xe and H₂-N₂ are borderline systems, even though one is a noble gas pair and the other has more reliable data than any other except for He-Ar, because at temperatures above 1000 K they have uncertainty limits corresponding to Group II. This is due to the relatively large amount of scatter in the Kr-Xe results from molecular-beam measurements. For H₂-N₂ the potential was determined by use of the combination rules (sec. 2.6, part c), not by direct molecular-beam measurements. Additional uncertainties arise for diatomic molecules which are not rigorously treated in the theoretical model.

The gas pairs of Group II have values of \mathcal{D}_{12} which have been consistently verified by several independent studies and by different apparatus. There are, however, four borderline systems in Group II. Three of these, He-O₂, H₂-Ne, and N₂-Ar, do not have as many reliable measurements as the other gas pairs of Group II. The fourth gas pair, H₂-Kr, is a borderline system because the uncertainty limits are estimated to be ± 4 percent at 90 K and ± 3 percent at 500 K, which are limits slightly greater than the levels specified for Group II. Another gas pair, He-CO, is included in Group II because it has diffusion characteristics similar to He-N₂, or almost identical values of \mathcal{D}_{12} .

The gas pairs of Group III have a relatively small number of reliable measurements of \mathcal{D}_{12} at about room temperature. At temperatures above 1000 K the values of \mathcal{D}_{12} have relatively large uncertainties because large discrepancies exist in the molecular-beam measurements used to calculate values of \mathcal{D}_{12} , or because the beam measurements have been obtained from only one laboratory. Usually there are two laboratory sources for beam results.

The uncertainties in the miscellaneous systems are rather variable, as can be seen from table 11.

The uncertainties for H₂O-CO₂ look peculiar, but the higher accuracy at high temperatures is due to the existence of data by the point-source method.

5.2. Correlation Parameters

This section gives the correlation parameters for values of \mathcal{D}_{12} as a function of temperature and of composition.

The diffusion coefficients were correlated as a function of temperature in accordance with the semi-empirical reference equations discussed in section 4.3. The empirical constants for eq (4.3-1)

are listed in table 12, and for eq (4.3-2) in table 13; there are seventy-four gas pairs in all. For tables 12 and 13 the values of \mathcal{D}_{12} were adjusted to refer to equimolar mixtures, with two exceptions. First, systems involving air refer to trace diffusion through a large excess of air (see sec. 2.1, part b). Even when direct measurements were available for air, most of the constants were generated from the corresponding values of \mathcal{D}_{12} for N₂ and O₂ according to Blanc's law. In this way more reliable data are used as the basis of the reference equations; the direct measurements were always compared to results by Blanc's law and found to be in agreement. Second,

TABLE 12. Correlation parameters of eq (4.3-1) for \mathcal{D}_{12}

System	10 ³ A	s	10 ⁻⁸ $\frac{\mathcal{D}_0}{k}$	S	S'	T Range	Group
	$\frac{\text{atm-cm}^2}{s(\text{K})^2}$		K	K	(K) ²	K	
³ He- ⁴ He	32.4	1.501	0.0448	-0.9630	1.894	1.74-10 ⁴	II
³ He- ⁴ He	0.156	1.636	—	—	—	14.4-90.0	II
He-Ne	25.41	1.509	.212	1.87	—	65-10 ⁴	I
He-Ar	15.21	1.552	.410	1.71	—	77-10 ⁴	I
He-Kr	10.61	1.609	1.42	-32.65	2036.	77-10 ⁴	I
He-Xe	7.981	1.644	4.02	-68.87	5416.	169-10 ⁴	I
He-H ₂	27.0	1.510	0.0534	—	—	90-10 ⁴	II
He-N ₂	15.8	1.524	.265	—	—	77-10 ⁴	II
He-CO	15.8	1.524	.265	—	—	77-10 ⁴	II
Ne-Ar	8.779	1.546	1.94	1.82	1170.	90-10 ⁴	I
Ne-Kr	8.520	1.555	6.73	20.4	—	112-10 ⁴	I
Ne-Xe	6.747	1.584	19.0	10.1	—	169-10 ⁴	I
Ar-Kr	5.346	1.556	13.0	47.3	—	169-10 ⁴	I
Ar-Xe	5.000	1.563	36.8	59.9	—	169-10 ⁴	I
Ar-H ₂	23.5	1.519	0.488	39.8	—	242-10 ⁴	II
Kr-Xe	2.933	1.608	128	52.7	—	169-10 ⁴	I
Kr-H ₂	18.2	1.564	1.69	26.4	—	77-10 ⁴	II
H ₂ -D ₂	24.7	1.500	0.0636	6.072	38.10	14-10 ⁴	II
H ₂ -N ₂	15.39	1.548	.316	-2.80	1067.	65-10 ⁴	I
H ₂ -CO	15.39	1.548	.316	-2.80	1067.	65-10 ⁴	I
N ₂ -CO	4.40	1.576	1.57	-36.2	3825.	78-10 ⁴	II

TABLE 13. Correlation parameters of eq (4.3-2) for \mathcal{D}_{12}

System	10 ³ A	s	S	T Range	Group	System	10 ³ A	s	S	T Range	Group
	$\frac{\text{atm-cm}^2}{s(\text{K})^2}$		K	K			$\frac{\text{atm-cm}^2}{s(\text{K})^2}$		K	K	
He-CH ₄	3.13	1.750	—	298-10 ⁴	III	N ₂ -O ₂	1.13	1.724	—	285-10 ⁴	III
He-O ₂	4.37	1.710	—	244-10 ⁴	II	N ₂ -H ₂ O	0.187	2.072	—	282-373	Misc.
He-air	3.78	1.729	—	244-10 ⁴	II	N ₂ -CO ₂	3.15	1.570	113.6	288-1800	II
He-CO ₂	3.31	1.720	—	200-530	II	N ₂ -SF ₆	1.66	1.590	119.4	328-10 ⁴	III
He-SF ₆	3.87	1.627	—	290-10 ⁴	III	CO-O ₂	1.13	1.724	—	285-10 ⁴	III
Ne-H ₂	5.95	1.731	—	90-10 ⁴	II	CO-air	1.12	1.730	—	285-10 ⁴	III
Ne-N ₂	1.59	1.743	—	293-10 ⁴	III	CO-CO ₂	0.577	1.803	—	282-473	III
Ne-CO ₂	1.07	1.776	—	195-625	Misc.	CO-SF ₆	1.76	1.584	139.4	297-10 ⁴	III
Ar-CH ₄	0.784	1.785	—	307-10 ⁴	III	O ₂ -H ₂ O	0.189	2.072	—	282-450	Misc.
Ar-N ₂	.904	1.752	—	244-10 ⁴	II		2.78	1.632	—	450-1070	Misc.
Ar-CO	.904	1.752	—	244-10 ⁴	III	O ₂ -CO ₂	1.56	1.661	61.3	287-1083	III
Ar-O ₂	.977	1.736	—	243-10 ⁴	III	O ₂ -SF ₆	2.65	1.522	129.0	297-10 ⁴	III
Ar-air	.917	1.749	—	244-10 ⁴	III	air-H ₂ O	0.187	2.072	—	282-450	Misc.
Ar-CO ₂	1.74	1.646	89.1	276-1800.	III		2.75	1.632	—	450-1070	Misc.
Ar-SF ₆	1.48	1.596	145.4	328-10 ⁴	III	air-CO ₂	2.70	1.590	102.1	280-1800	III
Kr-N ₂	0.653	1.766	—	248-10 ⁴	III	air-SF ₆	1.83	1.576	121.1	328-10 ⁴	III
Kr-CO	.653	1.766	—	248-10 ⁴	III	H ₂ O-CO ₂	9.24	1.500	307.9	296-1640	Misc.
Xe-H ₂	3.68	1.712	16.9	242-10 ⁴	III	CO ₂ -N ₂ O	0.281	1.866	—	195-550	III
Xe-N ₂	0.470	1.789	—	242-10 ⁴	III	CO ₂ -C ₃ H ₈	.177	1.896	—	298-550	Misc.
H ₂ -CH ₄	3.13	1.765	—	293-10 ⁴	III	CO ₂ -SF ₆	.140	1.886	—	328-472	III
H ₂ -O ₂	4.17	1.732	—	252-10 ⁴	III	H-He	14.2	1.732	—	275-10 ⁴	Misc.
H ₂ -air	3.64	1.750	—	252-10 ⁴	II	H-Ar	1.45	1.597	—	275-10 ⁴	Misc.
H ₂ -CO ₂	3.14	1.750	11.7	200-550	II	H-H ₂	11.3	1.728	—	190-10 ⁴	Misc.
H ₂ -SF ₆	7.82	1.570	102.3	298-10 ⁴	III	N-N ₂	1.32	1.774	—	280-10 ⁴	Misc.
CH ₄ -N ₂	1.00	1.750	—	298-10 ⁴	III	O-He	4.68	1.749	—	280-10 ⁴	Misc.
CH ₄ -O ₂	1.68	1.695	44.2	294-10 ⁴	III	O-Ar	0.751	1.841	—	280-10 ⁴	Misc.
CH ₄ -air	1.03	1.747	—	298-10 ⁴	III	O-N ₂	1.32	1.774	—	280-10 ⁴	Misc.
CH ₄ -SF ₆	1.10	1.657	69.2	298-10 ⁴	III	O-O ₂	1.32	1.774	—	280-10 ⁴	Misc.

systems involving dissociated gases have data that are obtained from measurements of a trace atom diffusing through a mixture, or from calculations of \mathcal{D}_{12} based on molecular-beam measurements. Since the uncertainties in both cases are greater than the composition dependence of \mathcal{D}_{12} , it was unnecessary to adjust these data to an equimolar composition.

In tables 12 and 13 the gas pairs are ordered as follows: (1) mixtures of noble gases with noble gases arranged according to atomic weight of the lighter component, (2) mixtures of noble gases with other gases arranged according to the atomic weight of the noble gas, (3) other mixtures arranged according to the molecular weight of the lighter component, and (4) dissociated gases. Except for ^3He - ^4He and H_2 - D_2 isotopic mixtures are not included, since the self-diffusion coefficient is merely proportional to the viscosity.

The results given in tables 12 and 13 cannot be extrapolated to low temperatures, for the form of eqs (4.3-1) and (4.3-2) is unsuitable when the long-range London dispersion energy dominates the interaction (see sec. 2.6, part a). In such a case, values of \mathcal{D}_{12} may be obtained from the classical asymptote, eq (4.3-3). The correlation constants for the classical asymptotes are given in table 14;

TABLE 14. Classical low-temperature asymptotic values of \mathcal{D}_{12} , eq (4.3-3)

System	10°A	C ^a	Λ^{*b}	ϵ/k^b
	$\frac{\text{atm}\cdot\text{cm}^2}{s(\text{K})^{1/6}}$	$e^2\alpha_0^5$		K
He-Ne	31.2	3.0	1.32	23.7
He-Ar	20.3	9.6	0.86	40.2
He-Kr	17.9	13	.80	39.0
He-Xe	15.6	19	.68	46.5
He-CH ₄	19.0	14	.89	37
He-N ₂	20.4	10	.96	31
Ne-Ar	8.26	20	.35	61.7
Ne-Kr	6.79	27	.28	69.8
Ne-Xe	5.84	38	.26	69.1
Ne-H ₂	30.1	8.2	1.35	34
Ne-N ₂	8.69	21	0.37	57
Ar-Kr	3.51	91	.14	145
Ar-Xe	2.93	130	.11	178
Ar-H ₂	19.5	28	.87	64
Ar-CH ₄	5.27	98	.22	130
Ar-N ₂	4.93	69	.21	107
Kr-Xe	2.00	190	.08	197
Kr-H ₂	17.1	40	.75	80
Kr-N ₂	3.91	96	.16	132
Xe-H ₂	15.1	58	.67	87
Xe-N ₂	3.29	140	.14	145
H ₂ -CH ₄	17.5	43	.82	68
H ₂ -N ₂	19.3	30	.87	62.9
CH ₄ -N ₂	5.54	100	.23	120

^a Dalgarno (1967).

^b Based on the 12-6 potential parameters for noble gas pairs and for H_2 - N_2 from van Heijningen et al. (1966, 1968), and for other gas pairs from Hirschfelder et al. (1954).

a total of twenty-four gas pairs are listed which have London dispersion constants available.

If estimates of \mathcal{D}_{12} are required outside the temperature range of a reference equation, then care must be taken when extrapolations are made. At temperatures greater than 10 000 K, extrapolations are safer to make than at very low temperatures because of the form of the equations. However, at elevated temperatures an extrapolation will neglect the effects of inelastic collisions and internal excitation of molecules. When extrapolations have to be made at lower temperatures, both the reference equation and the low-temperature asymptote

should be used to obtain two predictions of \mathcal{D}_{12} at a given temperature. The larger value calculated is the better estimate of \mathcal{D}_{12} . This procedure neglects quantum effects, and unfortunately asymptotic constants are only available for about one-third of the gas pairs with recommended data.

If values of \mathcal{D}_{12} are required at pressures not equal to 1 atm, then the reciprocal pressure relationship of \mathcal{D}_{12} is used, as discussed in section 2.2.

The values of \mathcal{D}_{12} can be adjusted to a non-equimolar composition by the method developed in section 4.2. The values of the constants of eq (4.2-1) are given in table 15, using the same order of listing as described above for tables 12 and 13. Included in table 15 are a number of gas pairs in which D_2 replaces H_2 . Omitted from this table are mixtures with dissociated gases and several systems for which the molecular weights of the gases are so close that the composition dependence is negligible. These systems are He- D_2 , Ar- CO_2 , N_2 - CO , N_2 - O_2 , CO - O_2 , CO -air, CO_2 - N_2O , and CO_2 - C_3H_8 . Table 15 is convenient for making rapid estimates of the composition dependence of \mathcal{D}_{12} , or for correcting data to a specific composition, reliable to within the uncertainties of the experimental measurements.

5.3. Deviation Plots

The experimental diffusion coefficients are compared with correlated values of \mathcal{D}_{12} , and deviations are presented in a series of graphs, figures 5 to 81. Their sequence is in general accordance with the listing of gas pairs in table 10. There are no deviation plots for the mixtures with dissociated gases and for several other gas pairs which have only meager data available. The deviation plots do not present all the data for a given gas pair; results obtained from miscellaneous experimental methods or published in graphical form have been omitted. Systematic trends in the deviation plots should not be taken too seriously, since the reference equation is not theoretically precise.

The deviation plots show general features of experimental values of \mathcal{D}_{12} as follows. First, the overall consistency of the data is rather good, although some reported values of \mathcal{D}_{12} show considerable scatter. Second, careful appraisal of the experimental data is necessary to obtain the most reliable estimate of \mathcal{D}_{12} . A random selection of a value of \mathcal{D}_{12} from the literature could easily yield a result with an uncertainty of 5 percent, even though the original article would probably claim much less. Third, the results by the closed-tube and two-bulb methods are more consistent than others, and show no evidence of any systematic disagreement. This can be illustrated by the results for He-Ar and H_2 - N_2 which are given in figures 6, 7, 18, and 19. Fourth, most values of \mathcal{D}_{12} at temperatures above 1000 K are available only indirectly, that is from molecular-beam measurements. Fifth, in the approximate temperature range of 500 to 1000 K the point-source method has provided almost all the reliable data. Sixth, results from gas-chromatography measurements only supplement results by other methods for the gas pairs listed in table 10; however, gas-chromatography measurements give the only reliable data for many other mixtures. Finally, it has obviously been difficult to make any diffusion coefficient measurement with an uncertainty less than 1 percent.

TABLE 15. Correlation parameters for the composition dependence of \mathcal{D}_{12} according to eq (4.2-1)

System	ζ	ϵ/k^a	a	b	System	ζ	ϵ/k^a	a	b
		K					K		
³ He- ⁴ He	1.0	10.2	0.031	0.26	Xe-H ₂	1.0	87	0.25	1.53
He-Ne	1.64	23.7	.098	.45	Xe-D ₂	1.0	87	.23	1.43
He-Ar	1.67	40.2	.18	1.17	Xe-N ₂	1.0	145	.10	0.56
He-Kr	1.65	39.0	.23	1.56	H ₂ -D ₂	1.0	33	.042	.12
He-Xe	1.78	46.5	.29	2.08	H ₂ -CH ₄	1.0	68	.15	.94
He-H ₂	1.0	18.4	.033	-0.11	H ₂ -N ₂	1.00	62.9	.17	.89
He-CH ₄	1.0	37	.14	.25	H ₂ -CO	1.0	61	.16	.88
He-N ₂	1.80	31	.17	1.22	H ₂ -O ₂	1.0	61	.16	.81
He-CO	1.0	34	.16	1.19	H ₂ -air	1.0	57	.16	.87
He-O ₂	1.0	34	.17	1.11	H ₂ -CO ₂	1.84	80	.21	1.33
He-air	1.0	31	.17	1.19	H ₂ -SF ₆	1.0	93	.33	2.33
He-CO ₂	1.0	44	.23	1.74	D ₂ -CH ₄	1.0	68	.11	0.81
He-SF ₆	1.0	51	.39	3.09	D ₂ -N ₂	1.00	62.9	.13	.76
Ne-Ar	1.2	61.7	.059	0.57	D ₂ -CO	1.0	61	.13	.74
Ne-Kr	1.01	69.8	.12	.87	D ₂ -O ₂	1.0	61	.13	.66
Ne-Xe	1.25	69.1	.17	1.31	D ₂ -air	1.0	57	.13	.74
Ne-H ₂	1.0	34	.10	0.26	D ₂ -CO ₂	1.84	80	.18	1.20
Ne-D ₂	1.0	34	.078	.16	D ₂ -SF ₆	1.0	93	.31	2.26
Ne-N ₂	1.0	57	.043	.65	CH ₄ -N ₂	1.0	120	.035	0.05
Ne-CO ₂	1.0	82	.081	.98	CH ₄ -O ₂	1.0	124	.038	.00
Ar-Kr	1.4	145	.051	.30	CH ₄ -air	1.0	120	.035	.05
Ar-Xe	1.8	178	.086	.57	CH ₄ -SF ₆	1.0	188	.12	.50
Ar-H ₂	1.73	64	.17	.85	N ₂ -H ₂ O	1.0	266	.020	-0.32
Ar-D ₂	1.73	64	.14	.74	N ₂ -CO ₂	1.0	132	.041	.38
Ar-CH ₄	1.0	130	.046	.02	N ₂ -SF ₆	1.0	154	.14	1.04
Ar-N ₂	1.0	107	.029	.10	CO-CO ₂	1.0	145	.041	0.38
Ar-CO	1.0	117	.029	.10	CO-SF ₆	1.0	169	.14	1.06
Ar-O ₂	1.0	118	.026	.15	O ₂ -H ₂ O	1.0	296	.033	-0.03
Ar-air	1.0	109	.029	.11	O ₂ -CO ₂	1.0	147	.037	.44
Ar-SF ₆	1.0	179	.12	1.07	O ₂ -SF ₆	1.0	171	.14	1.14
Kr-Xe	1.8	197	.039	0.33	air-H ₂ O	1.0	274	.020	-0.34
Kr-H ₂	1.0	80	.21	1.14	air-CO ₂	1.0	136	.040	.39
Kr-D ₂	1.0	80	.19	1.07	air-SF ₆	1.0	159	.14	1.06
Kr-N ₂	1.0	132	.066	0.28	H ₂ O-CO ₂	1.0	384	.060	0.34
Kr-CO	1.0	145	.066	.28	CO ₂ -SF ₆	1.0	222	.088	.60

^a Based on the 12-6 potential; parameters for noble-gas pairs and for H₂-N₂ from van Heijningen et al. (1966, 1968), and for others from Hirschfelder et al. (1954).

Detailed remarks on the deviation plots are as follows. A positive deviation means that an experimental value of \mathcal{D}_{12} is greater than a value calculated from the reference equation. All values have been corrected to equimolar composition. Each deviation point has been plotted with a precision greater than 0.1 percent by means of a Calcomp plotter (model 563). When a number in parentheses is placed by a point, then this number specifies the magnitude of the deviation (which happens to be greater than the ordinate scale). The abscissa usually covers the temperature range from 63 to 10 000 K, but lower temperatures appear on a few graphs for ³He-⁴He and H₂-D₂. Along the abscissa, hash marks (#) have been used to condense the temperature scale at elevated temperatures. Because of this break in the scale, points from similar sources are not connected by lines between 1000 and 10 000 K, as is done at lower temperatures.

For each gas pair the recommended reference equation for \mathcal{D}_{12} is given with the deviation plot. These equations are exactly the same as in tables 12 and 13. Occasionally, below an equation the parenthetical statement "(same as . . .)" appears. This means that the diffusion characteristics of two gas pairs are so similar that one equation is suitable for the correlation of the data of both. Results by the closed-tube method are noted as "Loschmidt tube." The values of \mathcal{D}_{12} calculated in this report from molecular-beam scattering experiments are referred to by one of the two laboratory sources, namely "Beam data of Amdur et al." for data from the Massachusetts Institute of Technology, and

"Beam data of Leonas et al." for data from the Moscow State University. There are parenthetical notes in the legend, some of which indicate the following: (1) a prime author whose results were available only as reported by others; (2) two-bulb apparatus which has been used to produce both "relative" values of \mathcal{D}_{12} and the usual "absolute" values (relative values of \mathcal{D}_{12} are obtained by calibrating the apparatus against a mixture with known \mathcal{D}_{12}); (3) standard deviations that indicate significant internal scatter, as published; (4) the type of radioactive species used in some experimental determinations; (5) the basis of some values of \mathcal{D}_{12} , for instance, mixture viscosity.

5.4. Detailed Remarks

This section presents the detailed remarks on the critical evaluation and the correlations of \mathcal{D}_{12} . Reasons are given for the assignment of a gas pair into a particular category of reliability. Enough information is reported to allow the recovery and the verification of the reference equations; most of this information is presented in tables 17, 18, and 20 to 25. In these tables the sources of data noted by an asterisk are for values of \mathcal{D}_{12} selected from large-scale graphs by eye. A selected value is a reasonable estimate in a small temperature region; that is, no published value of \mathcal{D}_{12} was considered extraordinarily superior to other available measurements.

The general order of the detailed remarks is as follows. The remarks are divided into four sections corresponding to the four reliability groups of table

10. The discussions contain the following information: (1) weights for values of \mathcal{D}_{12} used in the least-squares calculations of the reference equations, (2) intermolecular potentials obtained from molecular-beam experiments which were used to calculate \mathcal{D}_{12} at elevated temperatures, and (3) special comments.

For this section, most references are to be found in the Bibliography by author, and are not given at the end of the section.

a. Group I (Deviation Plots, Figs. 5 to 20)

Weights and Potentials. Similar weights were assigned to values of \mathcal{D}_{12} to correlate the data of Group I. The accurate measurements (at equimolar composition) of van Heijningen et al. (1966, 1968) were weighted unity; almost every other determination of \mathcal{D}_{12} was disregarded at temperatures below 400 K for the ten noble gas pairs and $\text{H}_2\text{-N}_2$. At 1000 and 10 000 K selected values, which were based on molecular-beam measurements, were weighted 1/5 and 1/10, respectively; these values are listed in table 17. These are logarithms of \mathcal{D}_{12} which were read from the large-scale graphs and used directly in the calculations. At 1000 K, the selected diffusion coefficients were obtained by extrapolation of \mathcal{D}_{12} calculated from beam results down to room temperature, and extrapolation of data between 295 and 400 K up to elevated temperatures. The selected values of \mathcal{D}_{12} were taken to lie between these two extrapolations. At 10 000 K, the selected values approximate the mean of \mathcal{D}_{12} based on the molecular-beam measurements of Amdur et al. and Leonas et al.

The above weighting policy had a few exceptions as follows. First, for He-Kr the weight of the datum at 295 K was increased from unity to two; otherwise

TABLE 17. Selected high-temperature points for curve-fitting, Group I

System	$\log_{10}[\mathcal{D}_{12}(\bar{x}=1/2)]$	
	1000 K	10 000 K
He-Ne	0.930	2.680
He-Ar	.785	2.555
He-Kr	.700	2.510
He-Xe	.630	2.435
Ne-Ar	.410	2.150
Ne-Kr	.320	2.065
Ne-Xe	.250	2.000
Ar-Kr	.070	1.810
Ar-Xe	.010	1.730
Kr-Xe	-0.150	1.600
$\text{H}_2\text{-N}_2$.790	2.575

the calculated deviations would have exceeded the uncertainty limits of Group I. An additional point at 77 K ($\mathcal{D}_{12}=0.0607$) was used in the least-squares calculations, and it was weighted 3/10. This value of \mathcal{D}_{12} was obtained from calculations based on the temperature dependence of the thermal diffusion factor by Annis et al. (1968) and normalized to the 295 K datum of van Heijningen et al. (1968). Second, for $\text{H}_2\text{-N}_2$ an additional datum at 562 K ($\log_{10} T=2.750$, $\log_{10} \mathcal{D}_{12}=0.365$) was included with a weight of 1/3 in the least-squares calculations. This point was used in order to improve the interpolation between the highest temperature (295 K) result by van Heijningen et al. (1966) and the selected point at 1000 K.

At temperatures greater than about 1000 K, values of \mathcal{D}_{12} were based on intermolecular potentials obtained from molecular-beam scattering experiments. Leonas et al. performed beam experiments for each of the ten noble gas pairs, and also determined

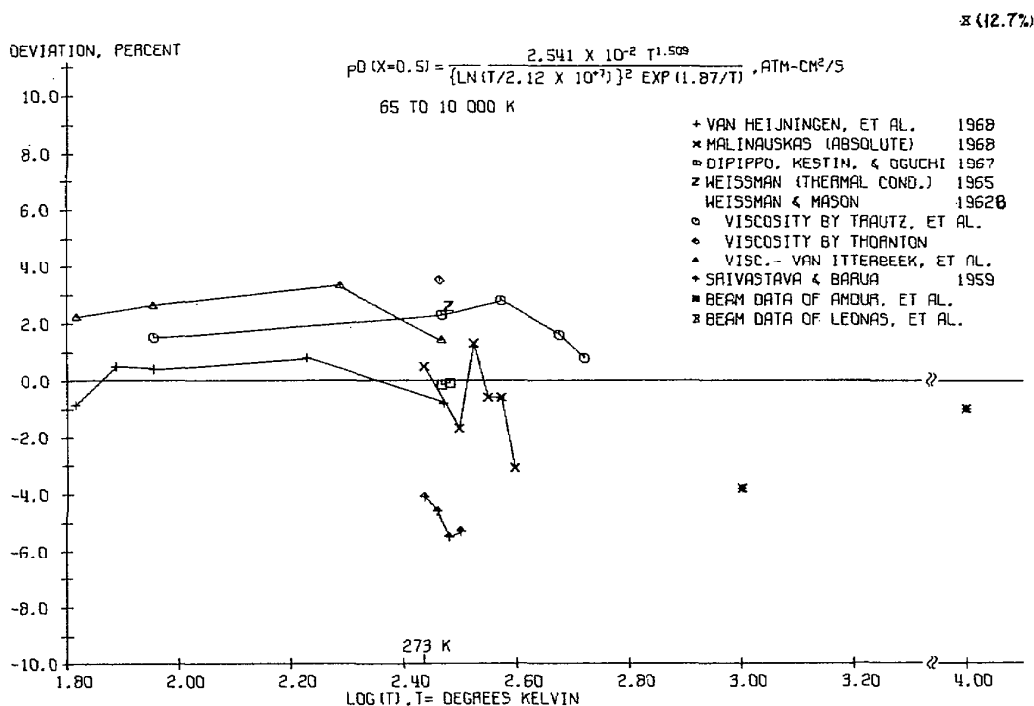


FIGURE 5. Deviations of diffusion coefficients from reference equation.

Helium-Neon

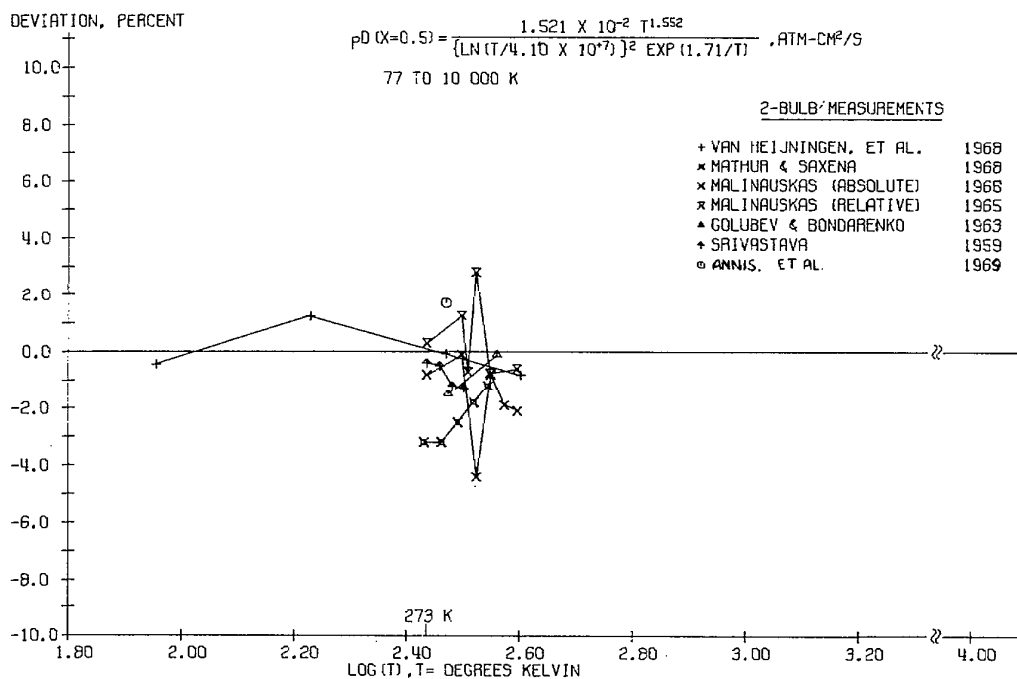


FIGURE 6. Deviations of diffusion coefficients from reference equation.

Helium-Argon

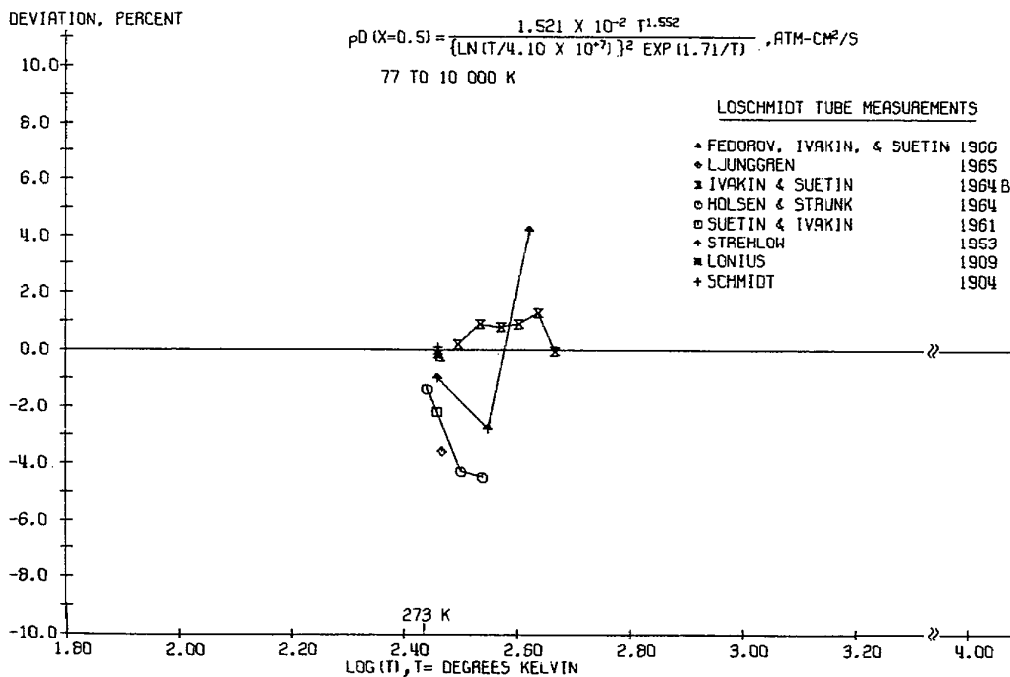


FIGURE 7. Deviations of diffusion coefficients from reference equation.

Helium-Argon

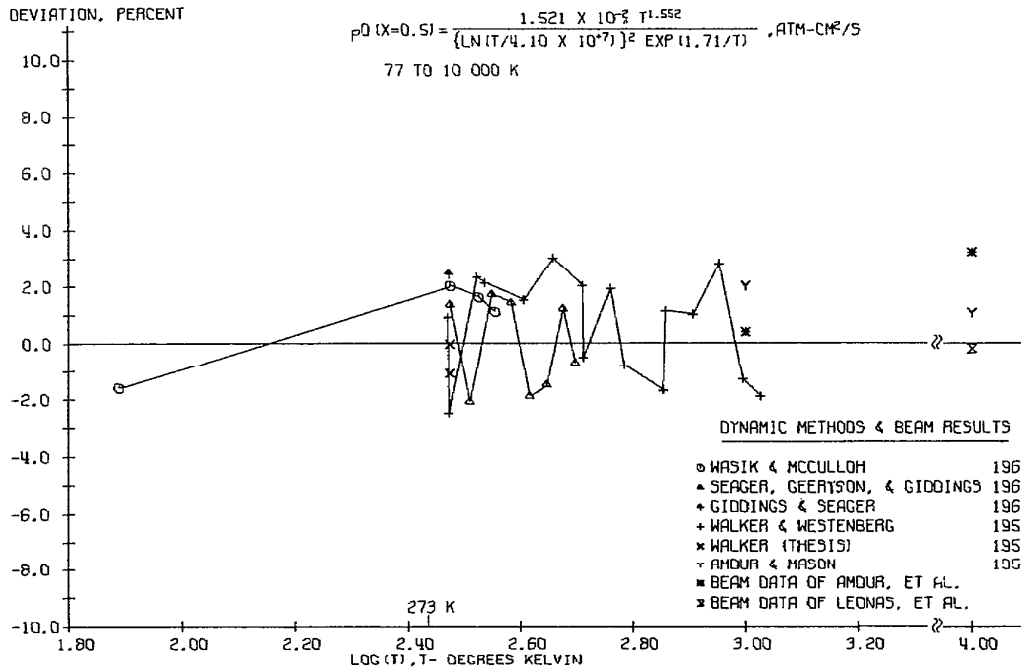


FIGURE 8. Deviations of diffusion coefficients from reference equation.

Helium-Argon

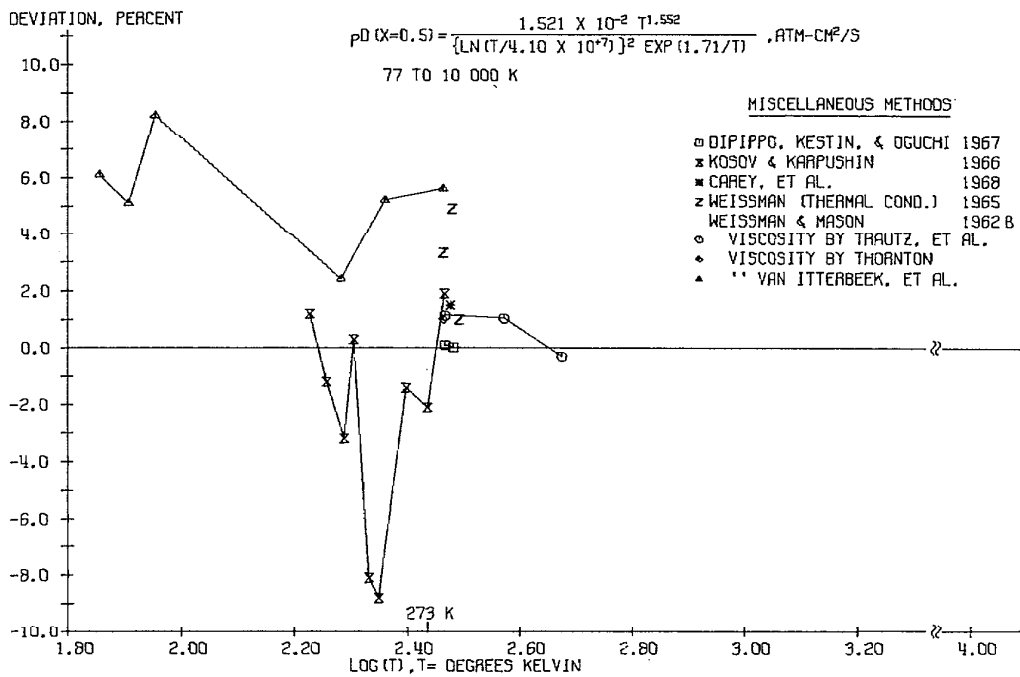


FIGURE 9. Deviations of diffusion coefficients from reference equation.

Helium-Argon

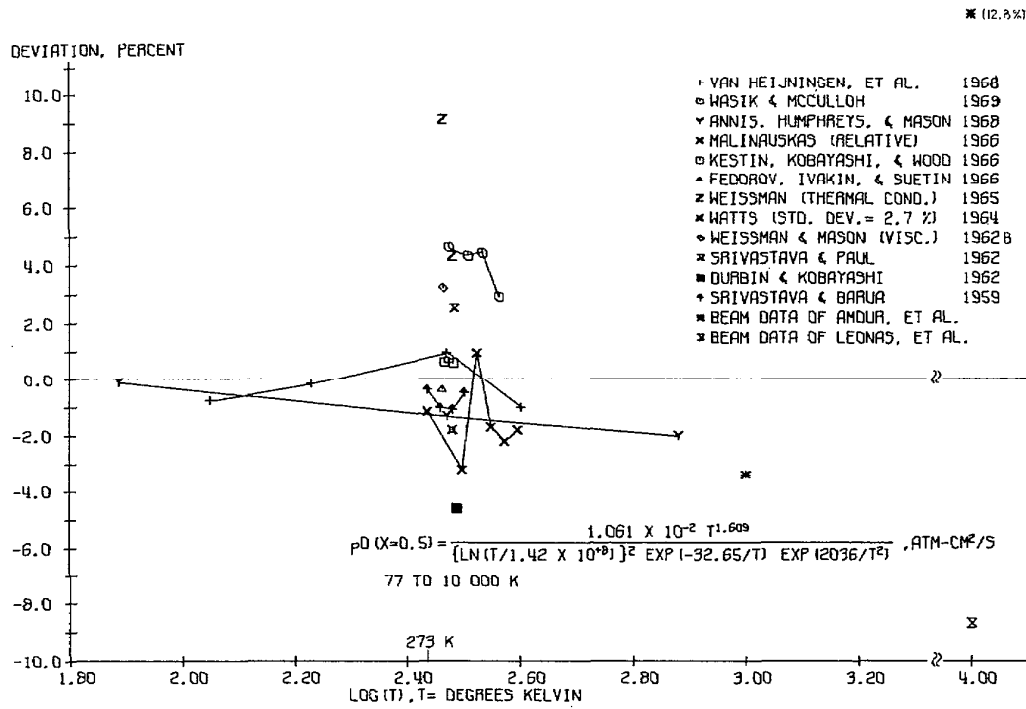


FIGURE 10. Deviations of diffusion coefficients from reference equation.

Helium-Krypton

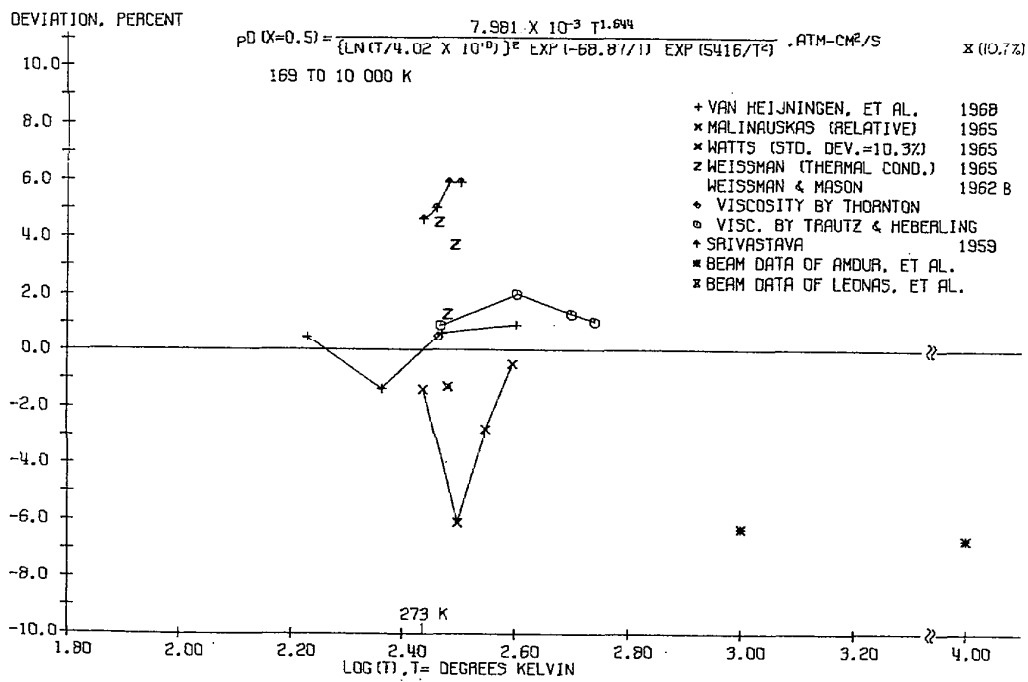


FIGURE 11. Deviations of diffusion coefficients from reference equation.

Helium-Xenon

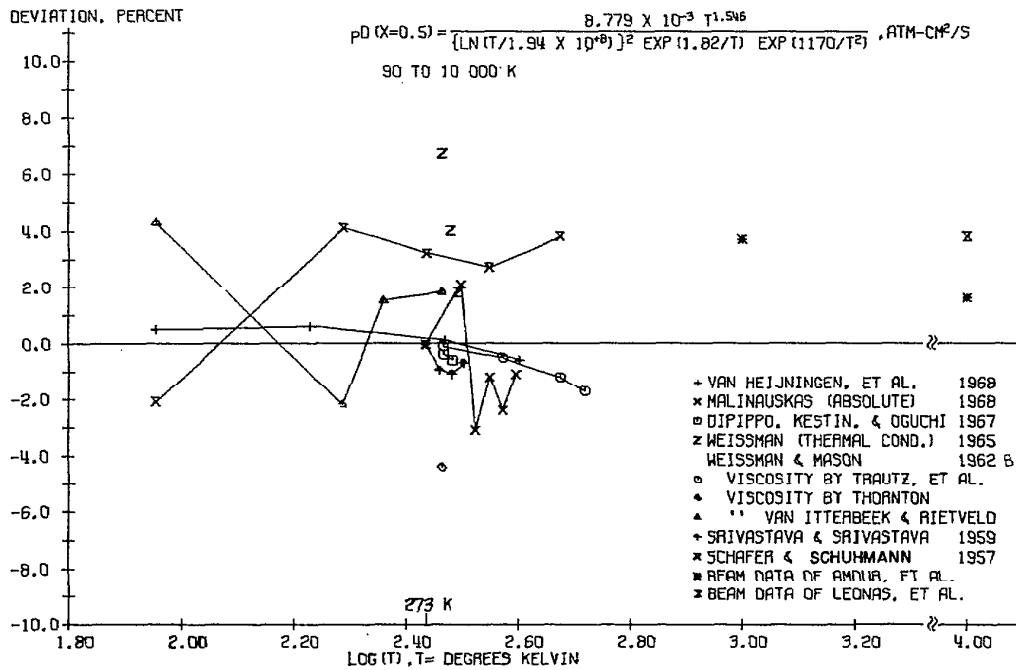


FIGURE 12. Deviations of diffusion coefficients from reference equation.

Neon-Argon

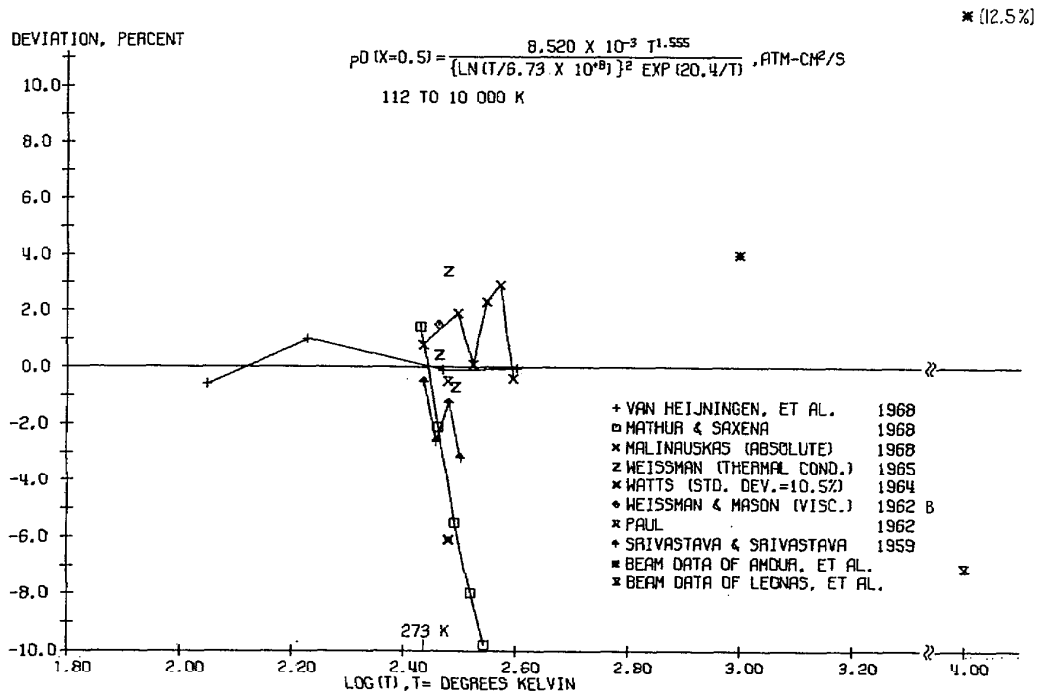


FIGURE 13. Deviations of diffusion coefficients from reference equation.

Neon-Krypton

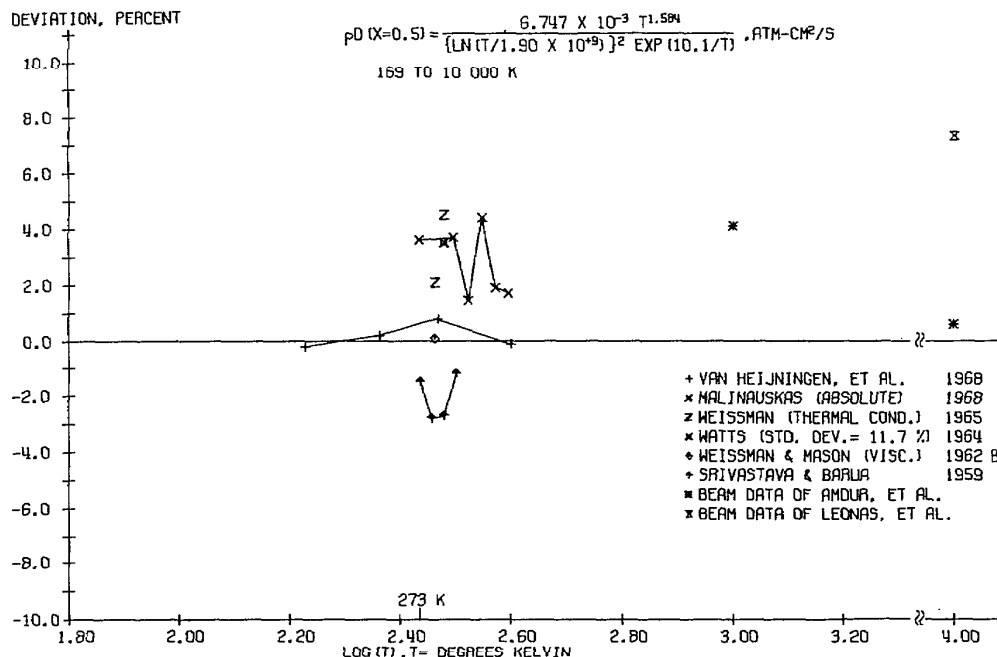


FIGURE 14. Deviations of diffusion coefficients from reference equation.

Neon-Xenon

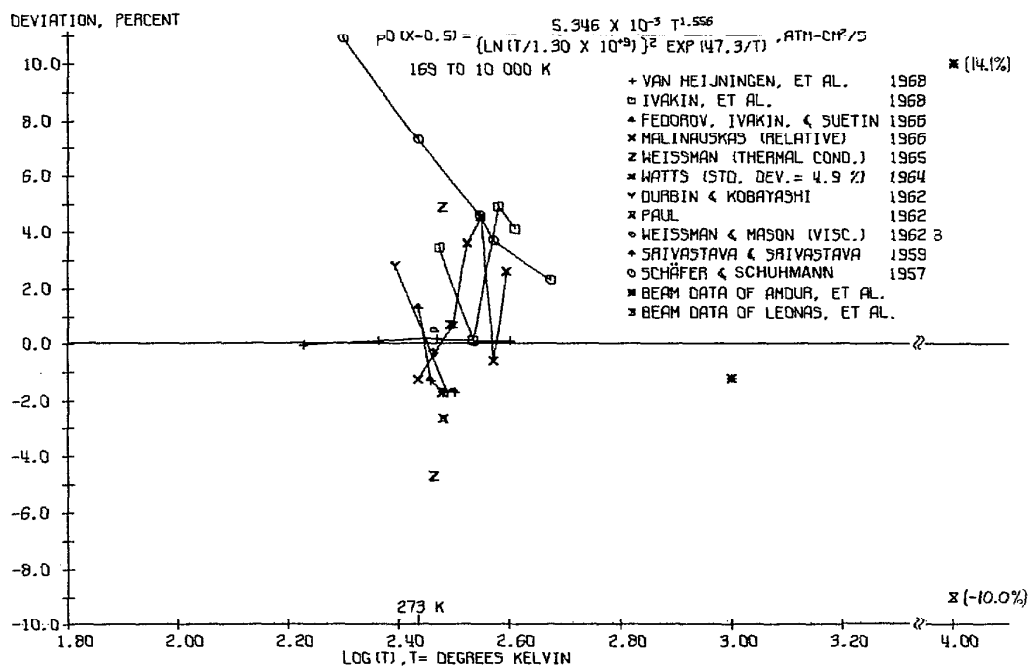


FIGURE 15. Deviations of diffusion coefficients from reference equation.

Argon-Krypton

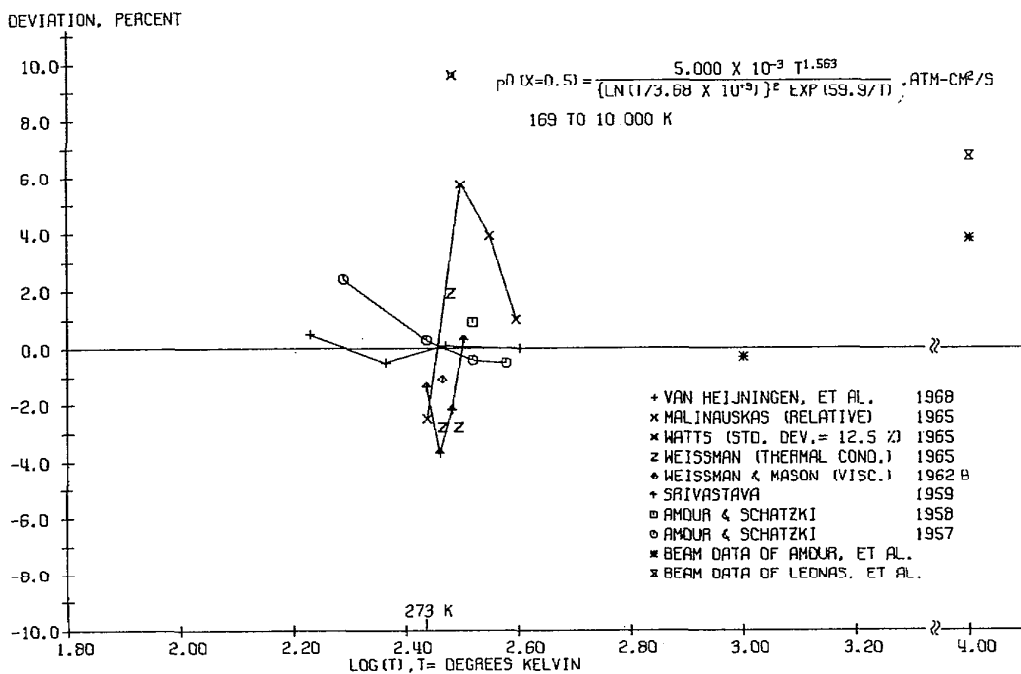


FIGURE 16. Deviations of diffusion coefficients from reference equation.

Argon-Xenon

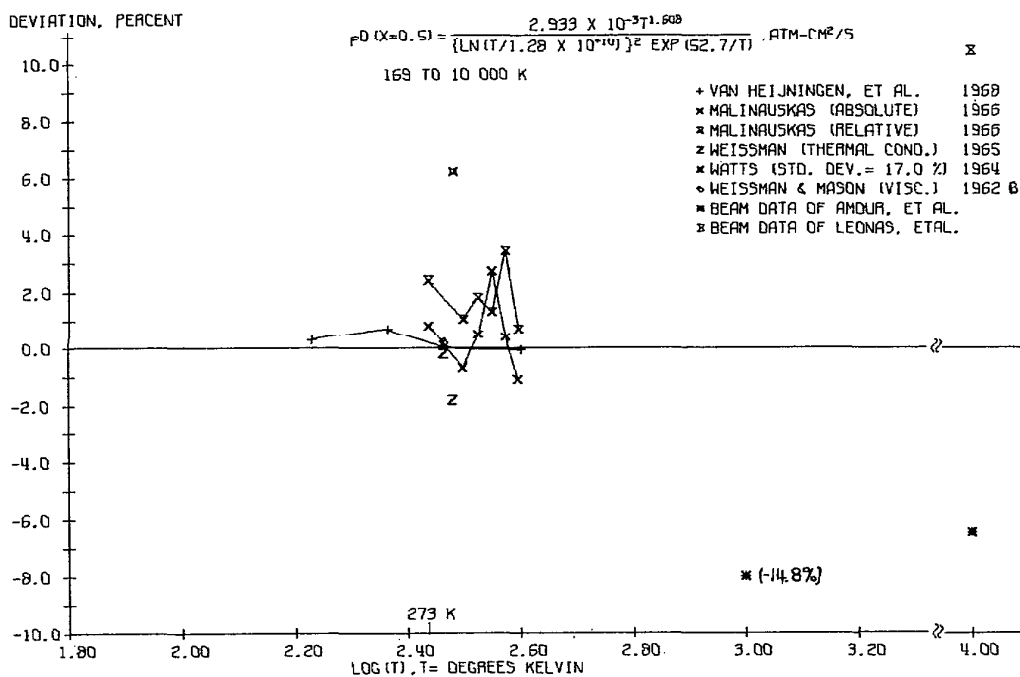


FIGURE 17. Deviations of diffusion coefficients from reference equation.

Krypton-Xenon

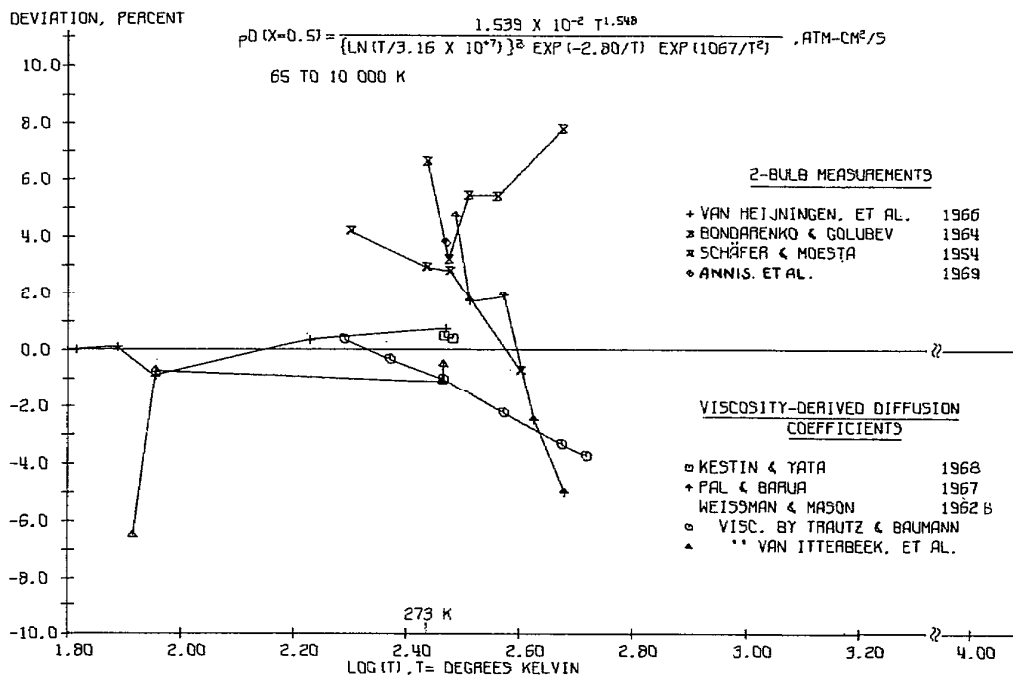


FIGURE 18. Deviations of diffusion coefficients from reference equation.

Hydrogen-Nitrogen

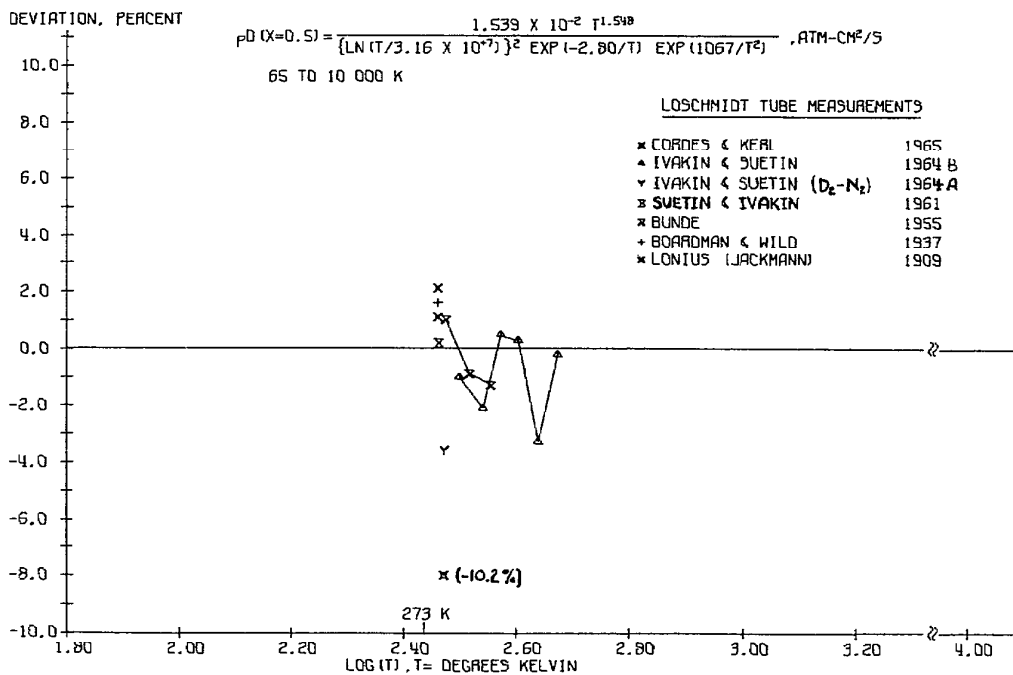


FIGURE 19. Deviations of diffusion coefficients from reference equation.

Hydrogen-Nitrogen

could not be found. The results by Srivastava are all below those by van Heijningen et al., except those for He-Xe, which are high. The lack of internal scatter in these measurements by Srivastava is presumably due to smoothing the data.

An interesting result for the gas pairs of Group I is that values of \mathcal{D}_{12} by direct measurements are in reasonable agreement with \mathcal{D}_{12} determined from other transport properties, as shown in table 19. Here direct measurements by van Heijningen et al. (1966, 1968) are compared with \mathcal{D}_{12} calculated from mixture viscosity and thermal conductivity data, reported by Weissman and Mason (1962 b) and by Weissman (1965). For table 19 the deviations were extracted from results given in the deviation plots for Group I. A relative index of reliability has also been computed, which is defined as the average absolute deviation of the results by van Heijningen et al. divided into the average absolute value of the other deviations, and it is given in the bottom row of table 19. These results indicate that \mathcal{D}_{12} can be well predicted from other transport property data at about room temperature. The diffusion coefficients calculated from the most accurate mixture viscosity data available (Kestin et al.) appear to be better than they should; that is, the viscosity-derived \mathcal{D}_{12} are really less reliable than the direct measurements of \mathcal{D}_{12} because the uncertainties in the A_{12}^* values are no less than 1 percent. The other mixture-viscosity sources yield \mathcal{D}_{12} only as reliable as the Group III uncertainty limits. However, diffusion coefficients calculated from available mixture thermal conductivities fall outside the range of Group III, or the average deviation is greater than 3 percent at about 300 K. This occurs because thermal conductivity measurements have much larger uncertainties than viscosity data, and not from any inadequacies of the theoretical formula.

b. Group II (Deviation Plots, Figs. 21 to 46)

Weights and Potentials. The equimolar values of \mathcal{D}_{12} and their weights used in the least-squares

calculations are presented in table 20. The potential functions obtained from molecular-beam measurements are summarized in table 21. A few systems have direct molecular-beam measurements, but most gas pairs of Group II have potentials that were obtained by the combination rules. The calculated potential functions are listed on the left-hand side of table 21, and the potentials from direct molecular-beam measurements are listed on the right-hand side. No potentials are given for He-CO₂, H₂-air, H₂-CO₂ and N₂-CO₂ because when this work was done, there were no molecular-beam measurements available with air or CO₂.

The potentials by Amdur et al. for He-CO, He-O₂, and H₂-CO were calculated by combination rules from measurements obtained in different apparatus. The potential energy ranges for these measurements were not the same. Thus, the derived potentials are applicable over a smaller temperature range, and are also considered slightly less reliable than results obtained from a single apparatus.

The potential for ³He-⁴He was taken the same as for ⁴He-⁴He, and that for H₂-D₂ the same as for H₂-H₂; that is, potentials were assumed identical for isotopic pairs. This is only an approximation, but is sufficiently accurate for the present purposes [1, 2].⁵

Special Comments. For the seventeen gas pairs of Group II special comments are as follows.

³He-⁴He. This gas pair is exceptional because its assignment into Group II is based mainly on the reliability of values of \mathcal{D}_{12} calculated from viscosity measurements. In some instances, viscosity data for ⁴He-⁴He (Becker and Misenta, 1955; Coremans et al., 1958 a; Rietveld et al., 1959) were used along with the appropriate reduced-mass correction factor and quantum-corrected values of A_{12}^* and $\Omega^{(1,1)*}$. At high temperatures values of \mathcal{D}_{12} were calculated from the ⁴He viscosity data by Kalekar and Kestin

⁵ Figures in brackets indicate the literature references at the end of Section 5.

TABLE 19. Values of \mathcal{D}_{12} by direct measurement compared with those from mixture viscosity, and from thermal conductivity

System	Direct measurement	Mixture viscosity				Thermal conductivity	
	van Heijningen et al.	Kestin	Trautz et al.	Thornton	van Itterbeek et al.	von Ubisch	Thornton
	(295 K)	(293 K)	(293 K)	(291 K)	(- 292 K)	(302 K)	(291 K)
Deviation from reference equations, percent							
He-Ne	-0.8	-0.15	+2.3	+3.5	+1.4	+2.6	+18.8
He-Ar	-0.05	+0.1	+1.2	+1.0	+5.6	+4.9	+3.3
He-Kr	+1.0	+0.6	—	+3.2	—	+4.3	+9.1
He-Xe	+0.6	—	+0.9	+0.5	—	+1.3	+4.5
Ne-Ar	+0.1	-0.4	-0.1	-4.4	+1.8	+4.0	+6.7
Ne-Kr	-0.1	—	—	+1.5	—	+3.4	+0.45
Ne-Xe	+0.8	—	—	+0.1	—	+4.5	+2.1
Ar-Kr	+0.2	—	—	-0.3	—	+4.9	-4.7
Ar-Xe	+0.1	—	—	-1.1	—	+1.9	-2.8
Kr-Xe	+0.05	—	—	+0.2	—	-1.8	-0.2
H ₂ -N ₂	+0.7	+0.5	-1.0	—	-0.8	—	—
[Avg. dev.]	0.41	0.35	1.1	1.6	2.4	3.4	5.3
Index of reliability	1	1	3	4	6	8	^a (3.8) 13 ^a (9)

^a Disregards large deviation (18.8%) of He-Ne.

TABLE 20. Diffusion coefficients and weights for curve-fitting, Group II

System	T, K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Weight	Note	System	T, K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Weight	Note		
${}^3\text{He}-{}^4\text{He}$	2.64	-3.1325	1/4	a	$\text{H}_2\text{-Ar}$	317	-.045	1	*		
	4.15	-2.8125	1/4	a		399	.140	1	*		
	1.74	-3.4789	1/4	b		501	.320	1	*		
	2.00	-3.3665	1/4	b		631	.495	1	*		
	2.31	-3.2396	1/4	b		794	.670	1	*		
	2.66	-3.1355	1/4	b		1000	.845	1	*		
	3.08	-3.0306	1/4	b		3170	1.710	1/3	*		
	3.96	-2.8386	1/4	b		10 000	2.590	1/6	*		
	14.4	-1.9066	1	b		$\text{H}_2\text{-Kr}$	77.0	-1.270	1/5	*, l	
	19.6	-1.7012	1	b			100	-1.040	1/5	*, l	
	64.8	-0.8327	1	b			178	-0.563	1/5	*, l	
	76.1	-.7282	1	b			290.7	-.1688	1	o	
	192	-.0742	1	b			296.0	-.1564	1	p	
	296	.2253	1	b			562	.334	1/5	*, l	
	290.15	.2550	1	c			3160	1.632	1/5	*, l	
	373.15	.4214	1	c			10 000	2.557	1/5	*	
	473.15	.5977	1	c			$\text{H}_2\text{-D}_2$	14.12	-2.3675	1	q
	573.15	.7405	1	c				15.47	-2.2832	1	q
	673.15	.8594	1	c		17.04		-2.1945	1	q	
	773.15	.9614	1	c		18.70		-2.1051	1	q	
	873.15	1.0527	1	c		20.32		-2.0329	1	q	
	1010.15	1.1550	1	c		90.0		-0.7721	1	q	
	1121.15	1.2307	1	c		26.09		-1.8097	1	r	
	2039	1.7127	1/4	d		32.57		-1.6091	1	r	
	7746	2.7774	1/4	d		41.35		-1.4117	1	r	
	3377	2.1038	1/4	e		48.06		-1.2832	1	r	
	10 000	2.9983	1/4	e		60.30	-1.1002	1	r		
2444	1.8639	1/4	f	70.32	-0.9851	1	r				
10 000	2.9908	1/4	f	200.0	-.1925	1	s				
10 000	2.9895	1/4	g	250.0	-.0292	1	s				
He-N_2	77.2	-1.1331	2	h	293.0	.0864	1	s			
	251	-0.265	1	*	400	.3181	1	s			
	317	-0.100	1	*	500	.4757	1	s			
	399	0.070	1	*	763	.7882	1	s			
	501	.241	1	*	986	.9741	1	s			
	631	.405	1	*	3313	1.9047	1	t			
	794	.575	1	*	5000	2.2305	1	t			
	1000	.745	1	*	10 000	2.7796	1	t			
	3170	1.640	2/5	*	$\text{H}_2\text{-CO}$	-	-	-	u		
	10 000	2.530	1/5	*		$\text{H}_2\text{-air}$	282	-0.1487	1	v	
He-CO	-	-	-	i	355		.0253	1	v		
	He-O_2	317	-0.085	1	*	447	.1987	1	v		
10 000		2.480	1	*	1000	.8048	1	v			
He-air	282	-0.1818	1	j	10 000	2.5635	1	v			
	355	-.0119	1	j	200.0	-0.5017	1	w			
	447	.1584	1	j	298.15	-.1898	1	x			
	1000	.7582	1	j	473.0	-.1673	1	y			
He-CO_2	10 000	2.4969	1	j	$\text{N}_2\text{-Ar}$	316	-.664	1	*		
	200	-0.5229	1	k		3160	1.088	1	*		
$\text{H}_2\text{-He}$	298.4	-.2240	1	l	$\text{N}_2\text{-CO}$	77.65	-1.7747	1/5	z		
	90.1	-.7012	1	a		194	-0.980	1/2	*		
	194.7	-.1264	1	m	251	-.790	1	*			
	251.2	.063	1	*	316	-.619	1	*			
	317	.232	1	*	398	-.450	1	*			
	399	.398	1	*	562	-.195	1/2	*			
	501	.567	1	*	1000	.226	1/5	*			
	1000	1.080	1/3	*	10 000	1.979	1/10	*			
	3170	1.970	1/3	*	$\text{N}_2\text{-CO}_2$	298.15	-0.7825	1	x		
	10 000	2.900	1/6	*		447	-.450	1/2	*		
$\text{H}_2\text{-Ne}$	90.1	-0.8416	1	a	708	-.095	1/2	*			
	9505	2.6599	1	n	1000	-.1553	1/2	aa			
$\text{H}_2\text{-Ar}$	251.2	-0.220	1	*	1800	.5832	1/2	aa			

* Selected value, see explanation in first part of section 5.4.

^a Weissman and Mason (1962 b).^b Bendt (1958).^c Calculated from viscosity data by Kalelkar and Kestin (1970).^d Calculated from molecular-beam potential by Amdur and Harkness (1954).^e Calculated from molecular-beam potential by Amdur et al. (1961 a).^f Calculated from molecular-beam potential by Belyaev and Leonas (1967 b).^g Calculated from molecular-beam potential by Kamnev and Leonas (1965 a).^h Wasik and McCulloh (1969).ⁱ Reference equation of He-N_2 is suitable because of isosteric molecules.^j Calculated from reference equations for He-N_2 and He-O_2 according to Blanc's law, eq (2.1-7).^k Calculated from temperature dependence of thermal diffusion factor (Saxena and Mason, 1959) according to the iterative method by Annis et al. (1968), and results are normalized to measurement of \mathcal{D}_{12} by Boyd et al. (1951).^l Annis et al. (1969).^m Amdur and Malinauskas (1965).ⁿ Calculated from molecular-beam potential by Amdur et al. see table 21.^o Fedorov et al. (1966).^p Annis et al. (1968).^q Calculated from HD viscosity data by Becker and Misenta (1955).^r Calculated from HD viscosity data by Coremans et al. (1958 b).^s Calculated from H_2 viscosity data as summarized by Mason and Rice (1954).^t Calculated from molecular-beam potential by Amdur et al. see table 21.^u Reference equation of $\text{H}_2\text{-N}_2$ is suitable because of isosteric molecules.^v Calculated from reference equations for $\text{H}_2\text{-N}_2$ and $\text{H}_2\text{-O}_2$ according to Blanc's law, eq (2.1-7).^w Calculated from temperature dependence of thermal diffusion factor (Saxena and Mason, 1959) according to the iterative method by Annis et al. (1968), and results are normalized to measurement of \mathcal{D}_{12} by Boyd et al. (1951).^x Boyd et al. (1951).^y Ivakin and Suetin (1964 b).^z Winn (1950).^{aa} Pakurar and Ferron (1966); Ferron (1967).

TABLE 21. Molecular-beam potentials, $\varphi(r) = K/r^s$, for Group II^{a,b}

System	Potential			Source			Reference
	K, eV(Å) ^s	s	Range, Å	System	K, eV(Å) ^s	s	
³ He- ⁴ He	4.71	5.94	1.27 -1.59	⁴ He- ⁴ He	4.71	5.94	Amdur and Harkness (1954).
He-N ₂	4.33	5.86	1.10 -1.53	⁴ He- ⁴ He	4.33	5.86	Belyaev and Leonas (1967 b).
	74.3	7.06	1.79 -2.29	Direct measurement			Amdur et al. (1957).
	48.8	6.63	1.72 -2.29	He-He	4.33	5.86	Belyaev and Leonas (1967 b).
He-CO	40.3	5.91	1.55 -2.26	N ₂ -N ₂	550	7.4	Belyaev and Leonas (1966 a).
	92.24	7.045	1.705-2.225	CO-Ar	551	6.99	Jordan et al. (1970).
				He-Ar	62.1	7.25	Amdur et al. (1954).
He-O ₂	9.5	7.26	1.47 -2.08	Ar-Ar	849	8.33	Amdur and Mason (1954).
	32.24	6.08	1.72 -2.34	He-He	4.33	5.86	Belyaev and Leonas (1967 b).
				CO-CO	1965	8.23	Belyaev et al. (1967).
H ₂ -He	12.11	6.07	1.44 -1.76	He-Ar	62.1	7.25	Amdur et al. (1954).
	5.0	3.8	1.15 -1.89	Ar-O ₂	1360	8.34	Jordan et al. (1970).
				Ar-Ar	849	8.33	Amdur and Mason (1954).
H ₂ -Ne	98.55	8.095	1.685-2.03	He-He	4.33	5.86	Belyaev and Leonas (1967 b).
	21	4.70	1.45 -2.215	He-Ar	62.1	7.25	Amdur et al. (1954).
				He-He	4.71	5.94	Amdur and Harkness (1954).
H ₂ -Ar	160	7.38	1.81 -2.44	He-He	4.71	5.94	Amdur and Harkness (1954).
	49.1	5.96 ₅	1.80 -2.54 ₅	H ₂ -H ₂	14.1	5.87	Belyaev and Leonas (1967 b).
				Ar-Ar	171	6.06	Kamnev and Leonas (1965 a).
H ₂ -Kr	70.37	5.81	2.01 ₅ -2.53 ₅	He-He	4.71	5.94	Amdur and Harkness (1954).
	89.33	4.72	1.80 -2.66	He-He	4.33	5.86	Belyaev and Leonas (1967 b).
				Kr-Kr	1382	7.7	Kamnev and Leonas (1966 a).
H ₂ -D ₂	31.55	6.19	1.62 -1.96	He-He	4.33	5.86	Belyaev and Leonas (1967 b).
	14.1	5.87	1.34 -1.95	He-He	12.11	6.07	Amdur and Smith (1968).
H ₂ -CO	107.4	5.81	1.89 -2.17	He-He	12.11	6.06	Amdur and Smith (1968).
	166.4	7.05	1.82 ₅ -2.43 ₅	He-D ₂	12.27	6.06	Amdur and Smith (1968).
				He-He	4.71	5.94	Amdur and Harkness (1954).
N ₂ -Ar	755	7.78	2.28 -2.83	H ₂ -H ₂	14.1	5.87	Belyaev and Leonas (1967 b).
	1050	8.16	2.12 -2.67	Ar-CO	551	6.99	Jordan et al. (1970).
				He-He	12.11	6.07	Amdur and Smith (1968).
N ₂ -CO	2038	8.70	2.22 -2.77	He-Ar	62.1	7.25	Amdur et al. (1954).
	596	7.27	2.43 -3.07	H ₂ -H ₂	14.1	5.87	Belyaev and Leonas (1967 b).
				CO-CO	1965	8.23	Belyaev et al. (1967).
				Direct measurement			Amdur et al. (1957).
				Direct measurement			Belyaev and Leonas (1967 a).
				Direct measurement			Belyaev et al. (1967).
				Direct measurement			Amdur et al. (1957).
				N ₂ -N ₂	596	7.27	Amdur et al. (1957).

^a Potentials were not determined for air-(He,H₂) and CO₂-(He,H₂,N₂) because molecular-beam measurements were unavailable.

^b Complete reference information is given in Bibliography II.

(1970). The direct measurements of \mathcal{D}_{12} by Bendt (1958) are in good agreement with the low-temperature results calculated from viscosity. There are other direct measurements for ³He-⁴He by DuBro (1969), which are not shown on the deviation plots because they were unavailable until recently. DuBro used a two-bulb method and covered the temperature range of 76.5 to 344 K. The average absolute deviation of his results from the reference equation is 2.6 percent.

In the temperature range of 14.4 to 90 K the form of eq (4.3-1) was not sufficiently flexible for curve-fitting purposes; that is, the data (obtained from mixture viscosities) were considered more accurate than the uncertainty specified for a Group II system in that temperature range. For temperatures between 14.4 and 90 K a simple power function was calculated by the method of least squares, in which

27 points were weighted equally. The result fits the data with an average absolute deviation of 1.1 percent and a standard deviation of 1.4 percent.

He-N₂. This gas pair has many reliable measurements by different major experimental methods which allow it to be a Group II system, see figures 23 and 24.

He-CO and H₂-CO. The diffusion coefficients of He-CO and H₂-CO can be well approximated by those for He-N₂ and H₂-N₂ (Group I), respectively, because CO and N₂ are isosteric molecules and He-N₂ and H₂-N₂ have more reliable measurements than do He-CO and H₂-CO. A comparison of the reference equations with the reliable measurements by Ivakin and Suetin (1964 a, b) for He-CO and H₂-CO shows deviations less than about 2 percent. The H₂-CO system was not assigned to Group I, as is H₂-N₂, because of possible uncertainties due to the

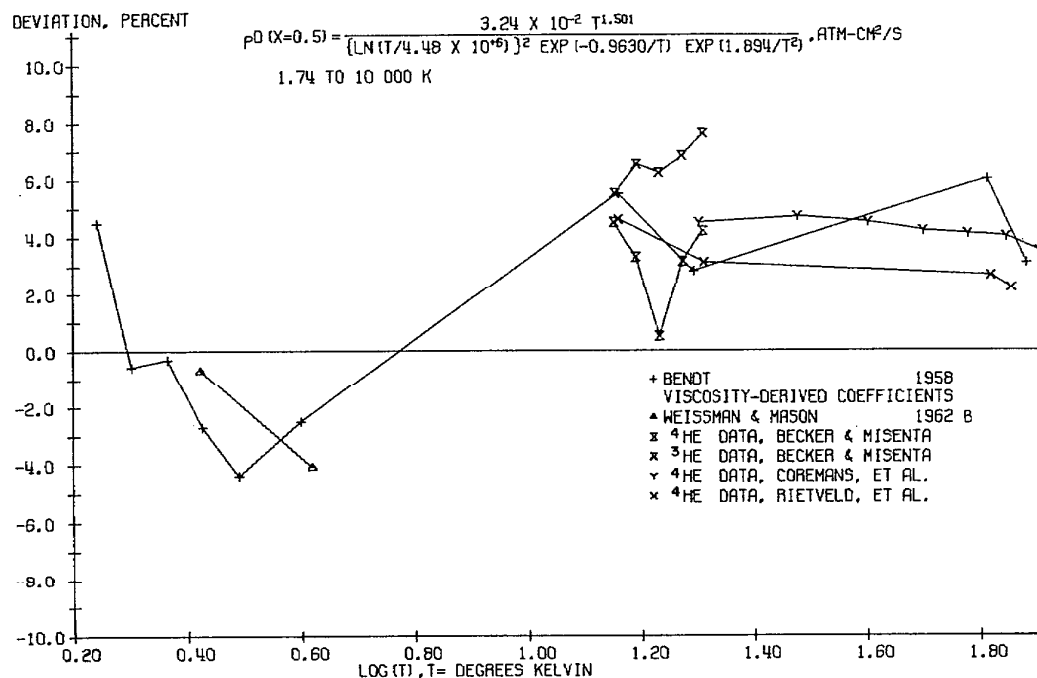


FIGURE 21. Deviations of diffusion coefficients from reference equation.

Helium-3-Helium-4

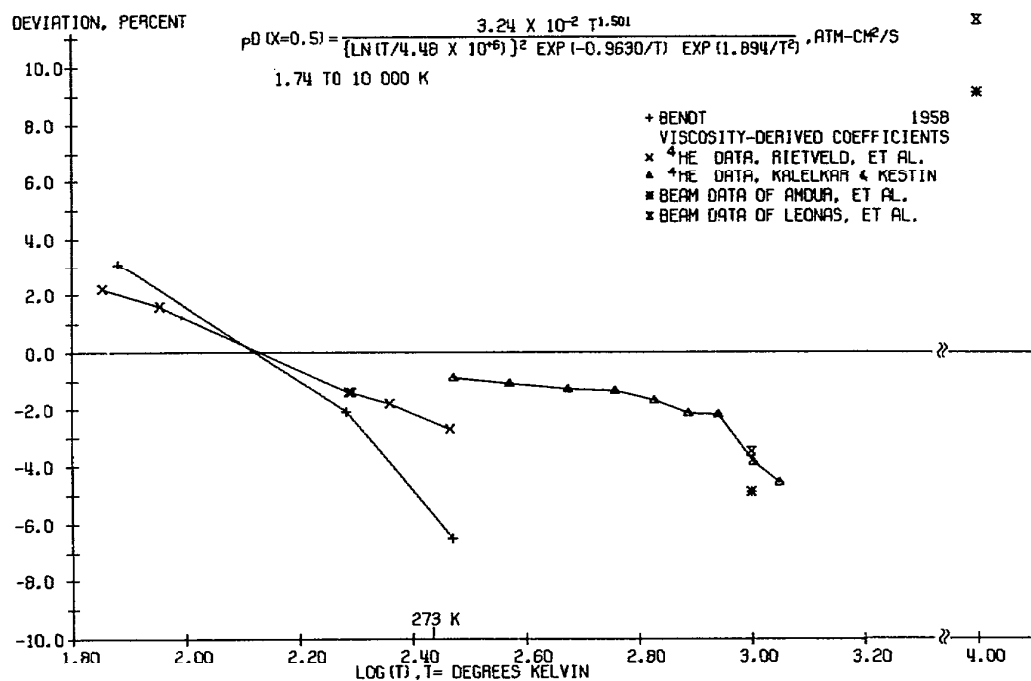


FIGURE 22. Deviations of diffusion coefficients from reference equation.

Helium-3-Helium-4

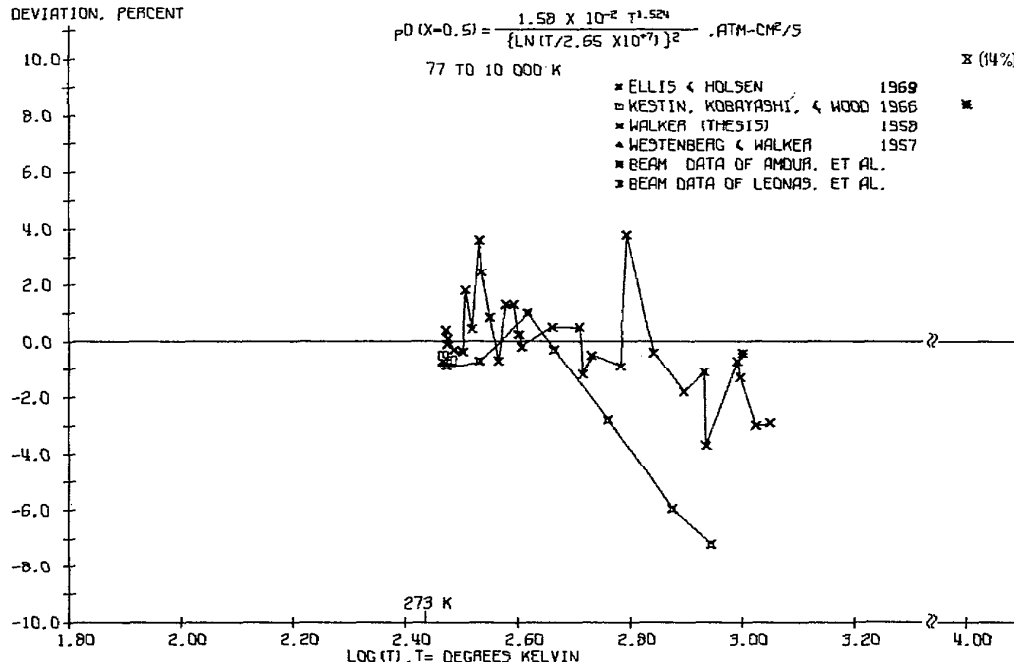


FIGURE 23. Deviations of diffusion coefficients from reference equation.
Helium-Nitrogen

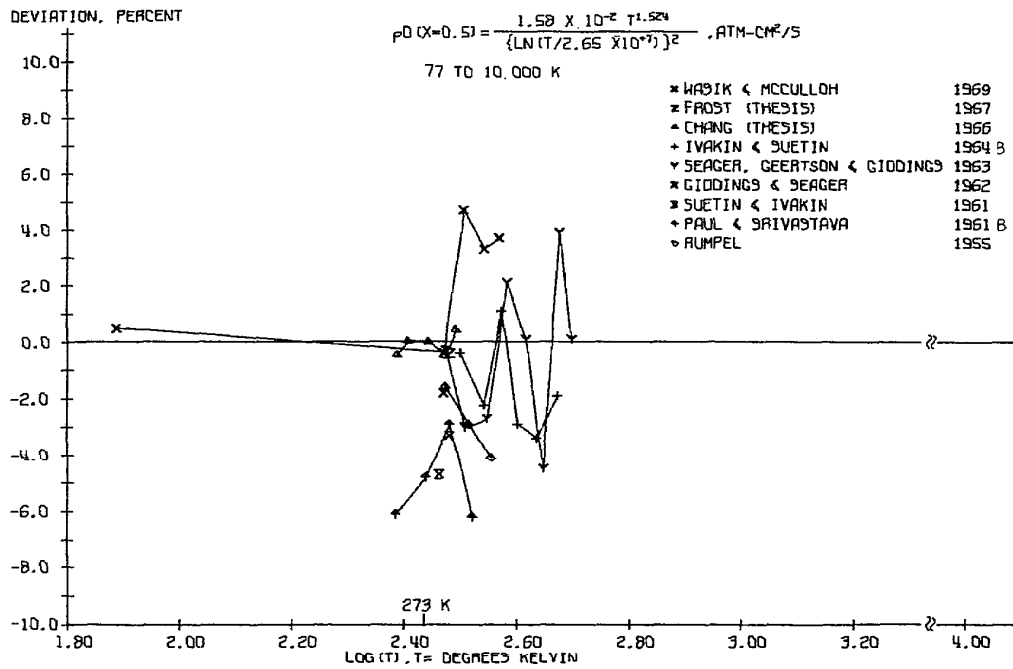


FIGURE 24. Deviations of diffusion coefficients from reference equation.
Helium-Nitrogen

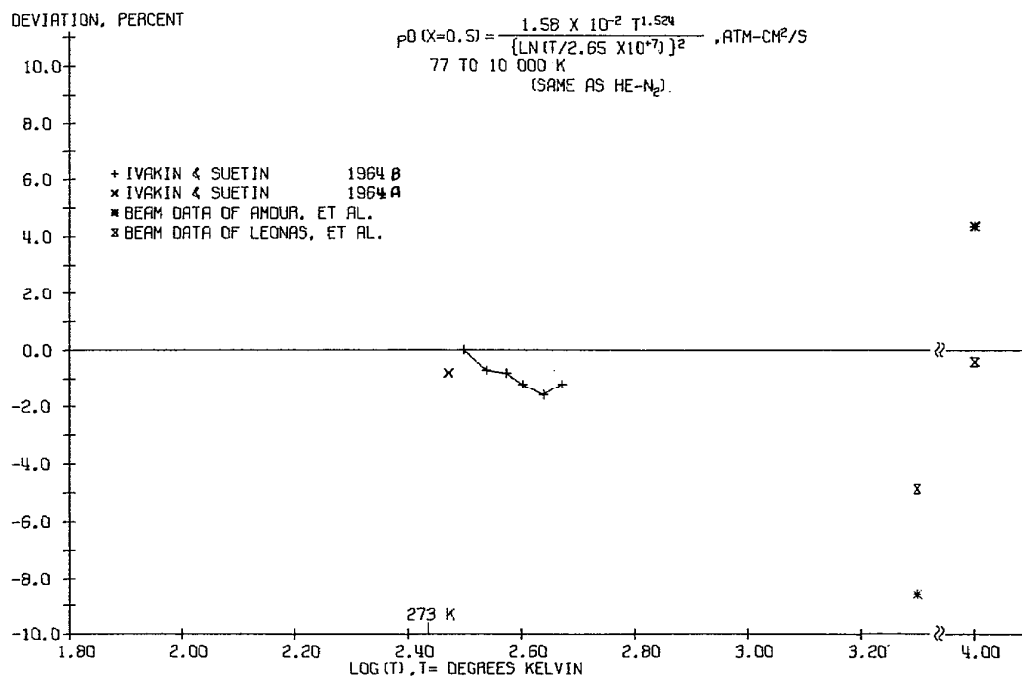


FIGURE 25. Deviations of diffusion coefficients from reference equation.

Helium-Carbon monoxide

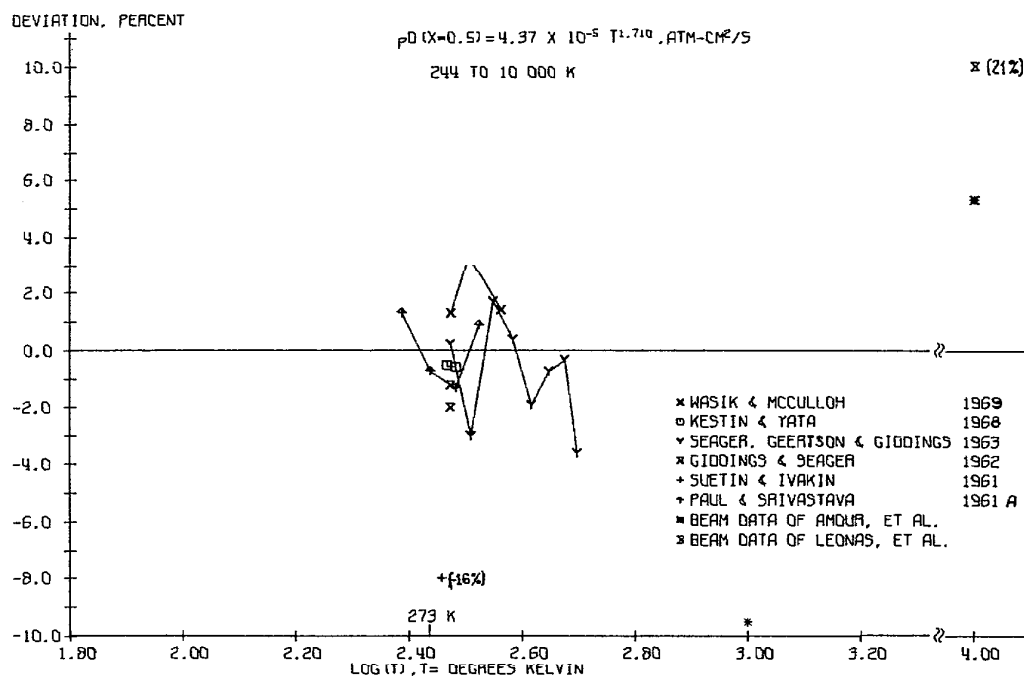


FIGURE 26. Deviations of diffusion coefficients from reference equation.

Helium-Oxygen

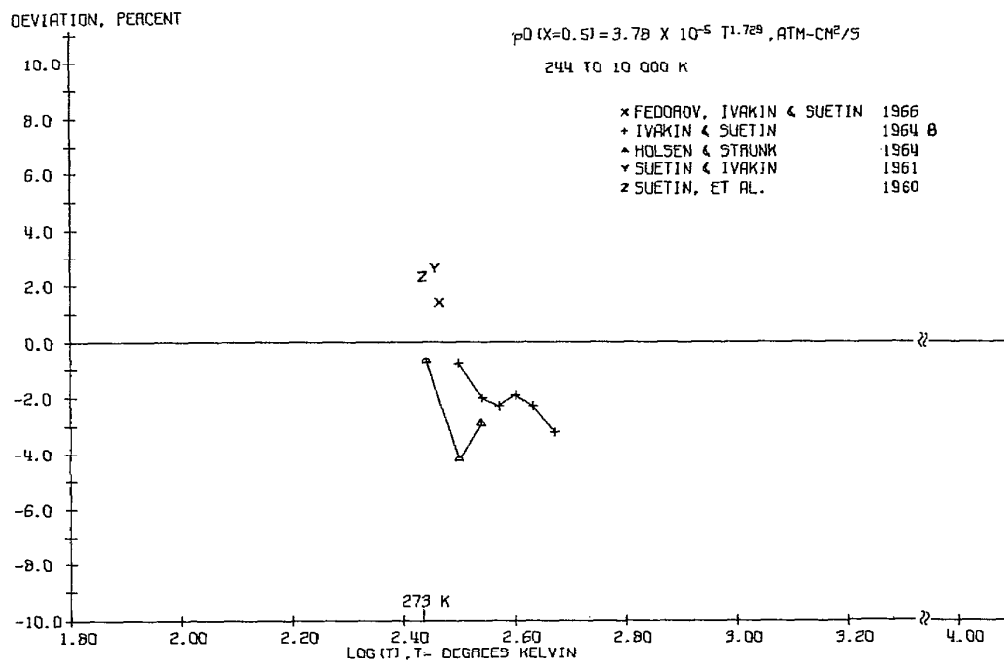


FIGURE 27. Deviations of diffusion coefficients from reference equation.
Helium-Air

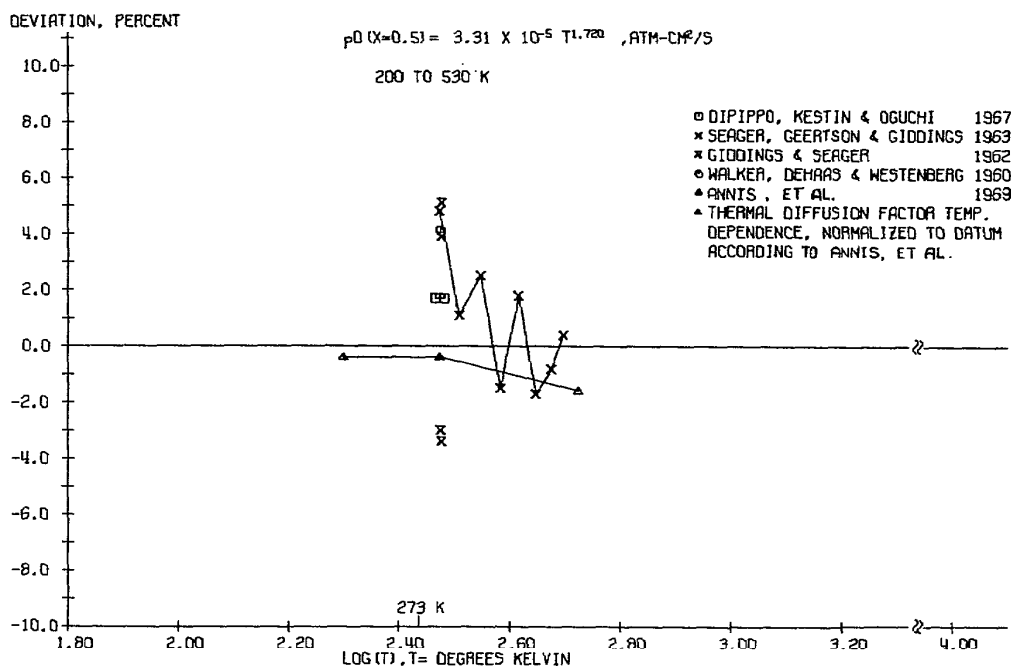


FIGURE 28. Deviations of diffusion coefficients from reference equation.
Helium-Carbon dioxide

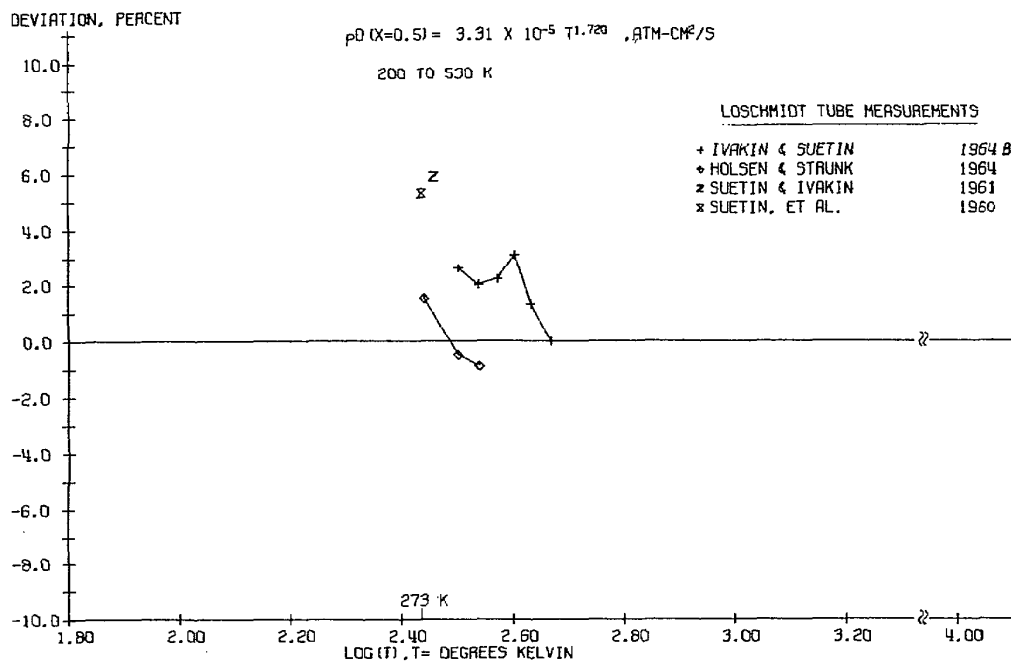


FIGURE 29. Deviations of diffusion coefficients from reference equation.

Helium-Carbon dioxide

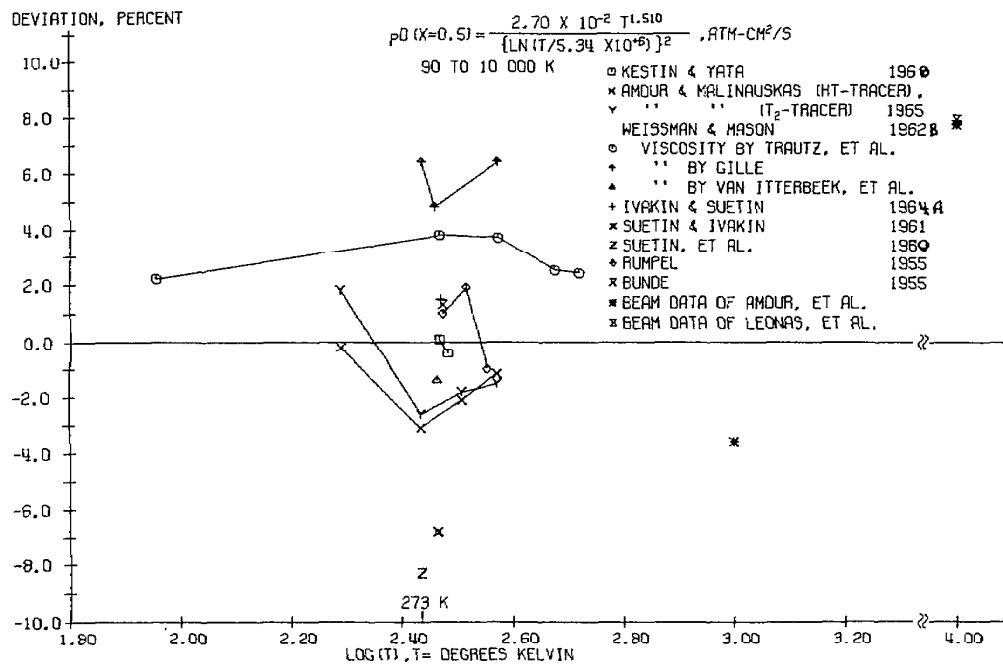


FIGURE 30. Deviations of diffusion coefficients from reference equation.

Hydrogen-Helium

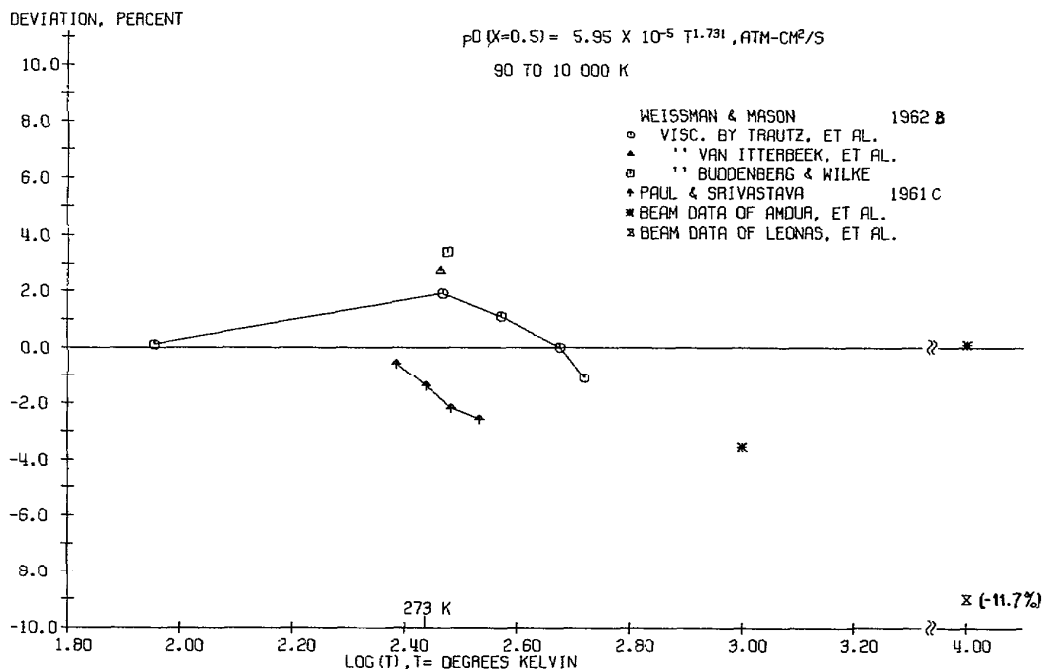


FIGURE 31. Deviations of diffusion coefficients from reference equation.

Hydrogen-Neon

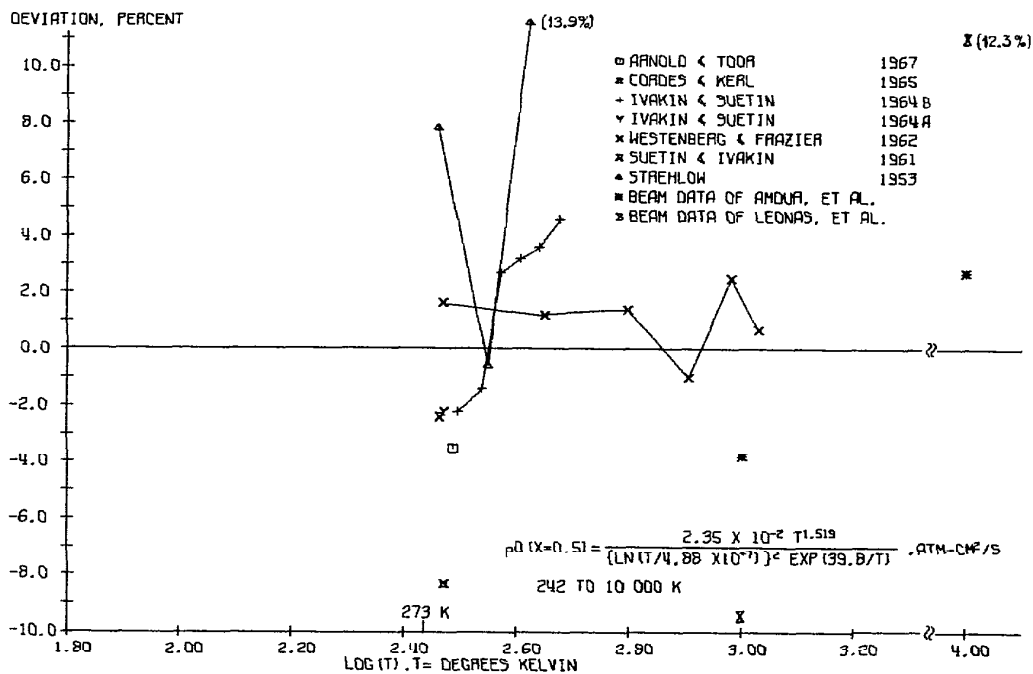


FIGURE 32. Deviations of diffusion coefficients from reference equation.

Hydrogen-Argon

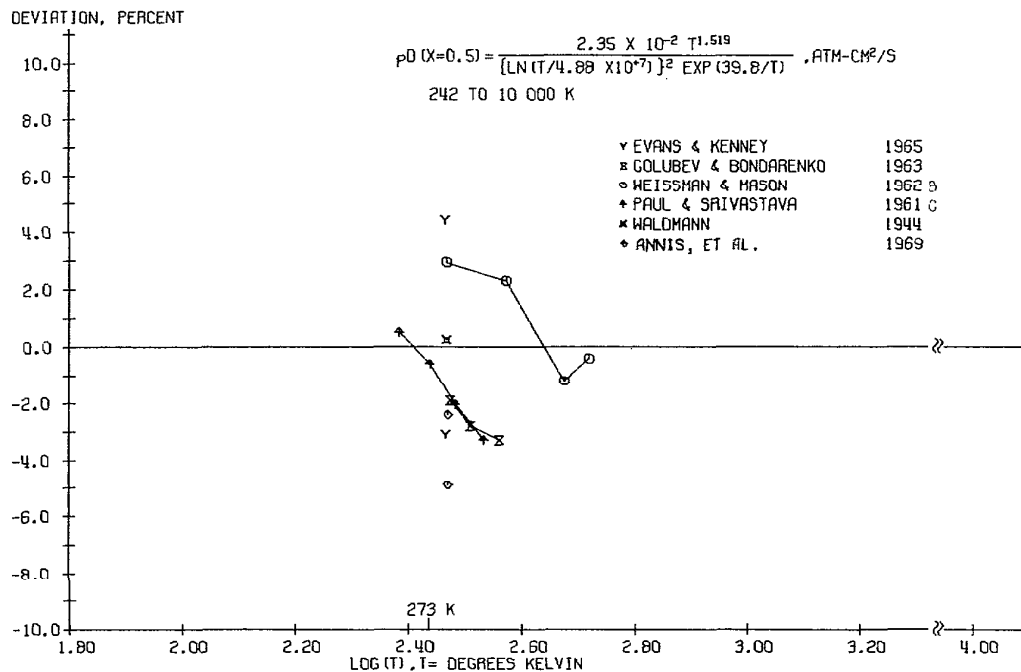


FIGURE 33. Deviations of diffusion coefficients from reference equation.

Hydrogen-Argon

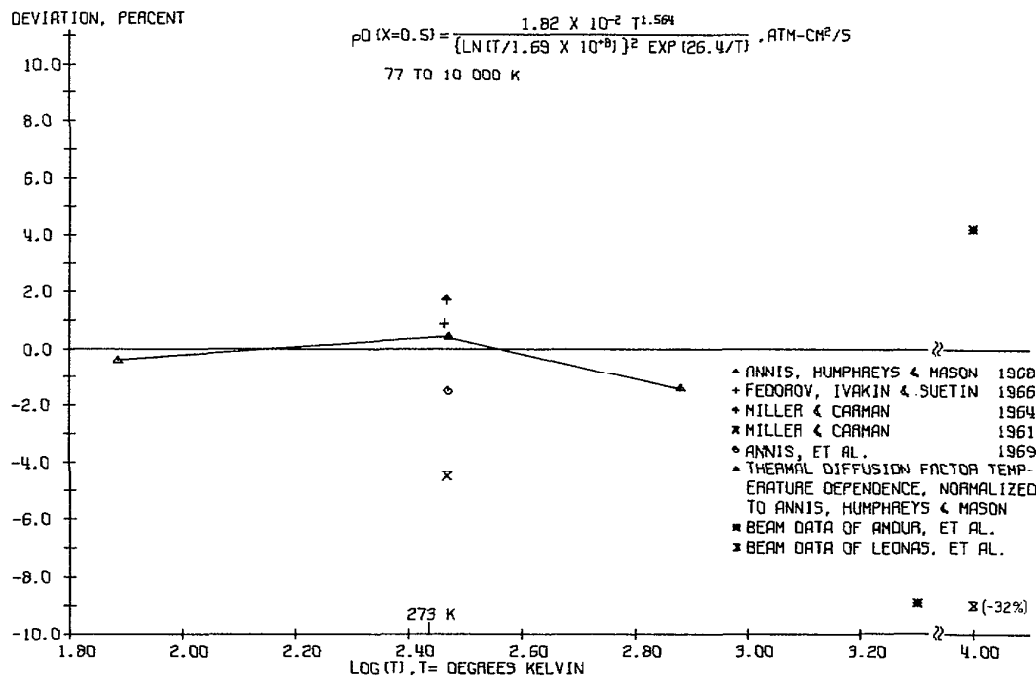


FIGURE 34. Deviations of diffusion coefficients from reference equation.

Hydrogen-Krypton

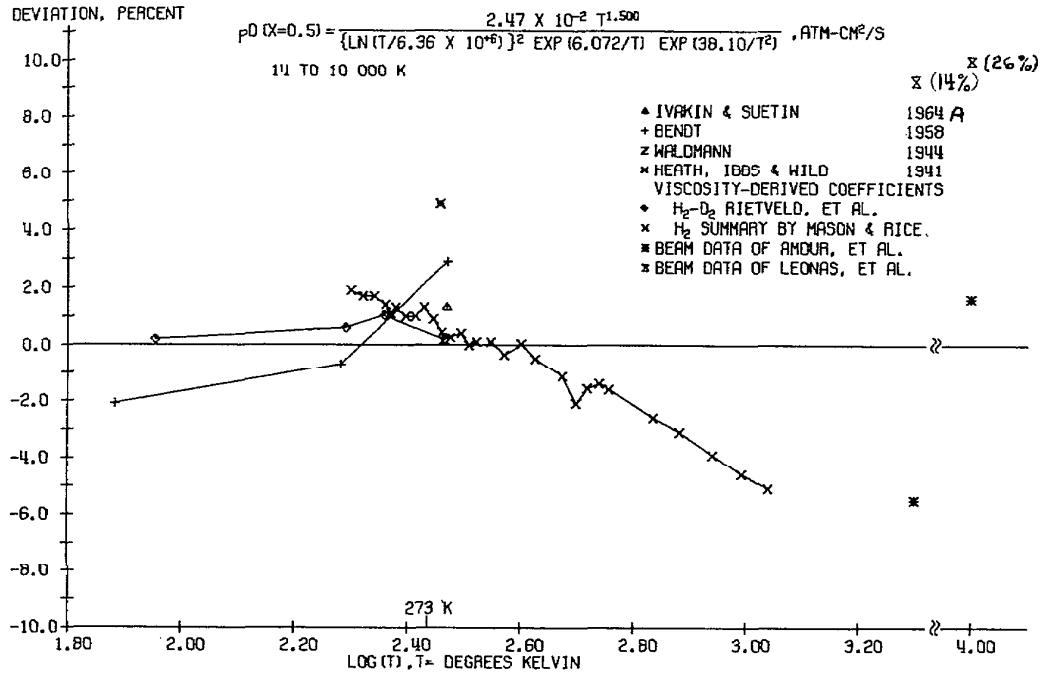


FIGURE 35. Deviations of diffusion coefficients from reference equation.
Hydrogen-Deuterium

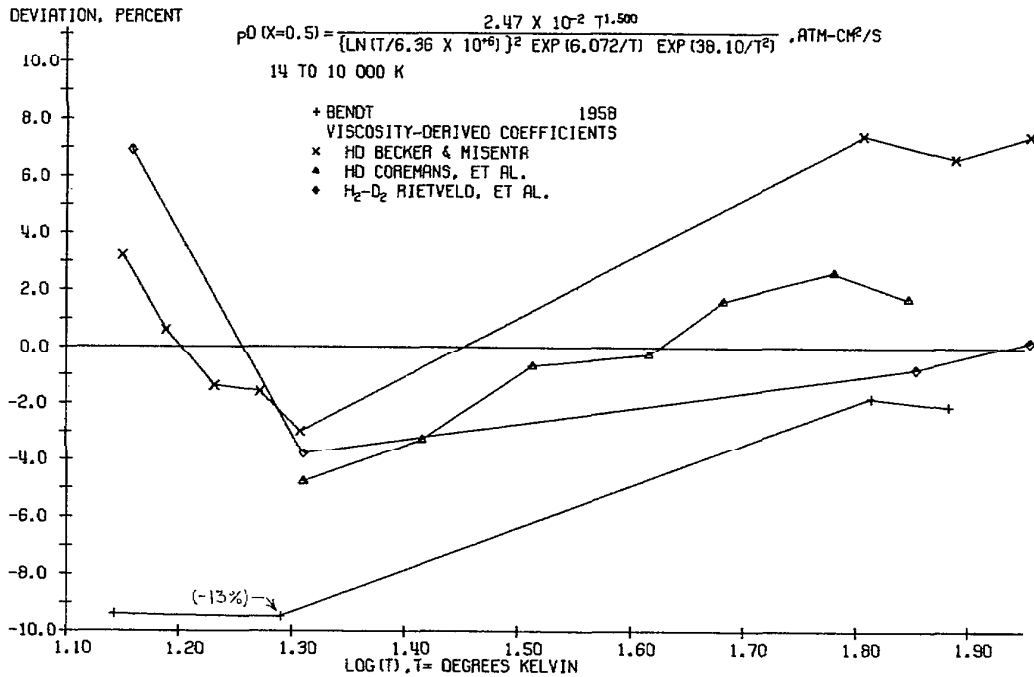
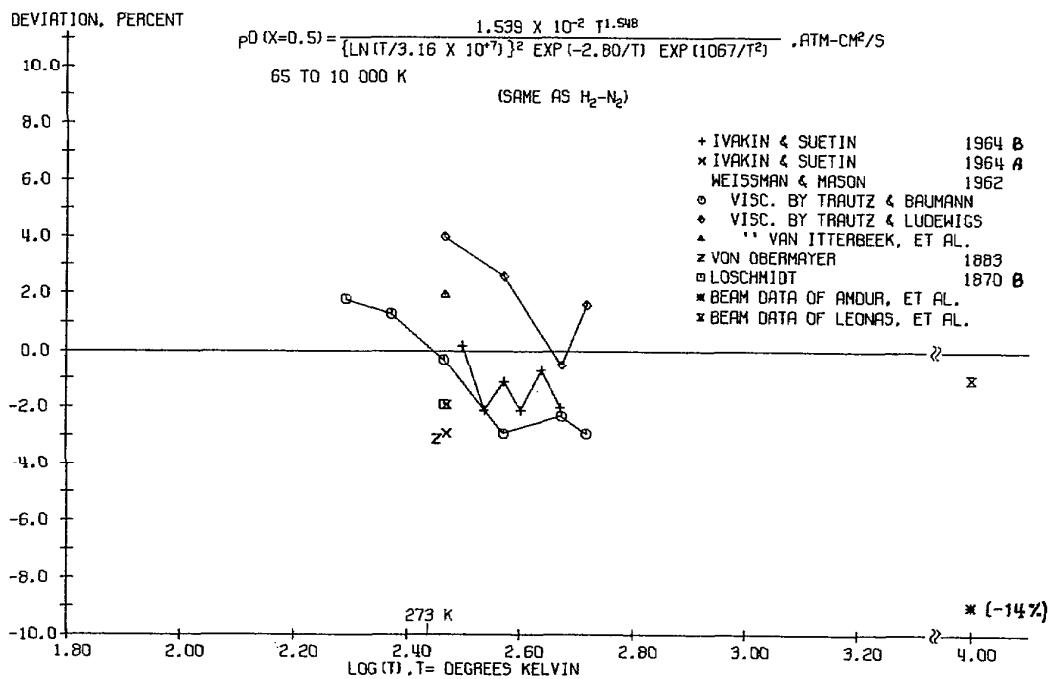
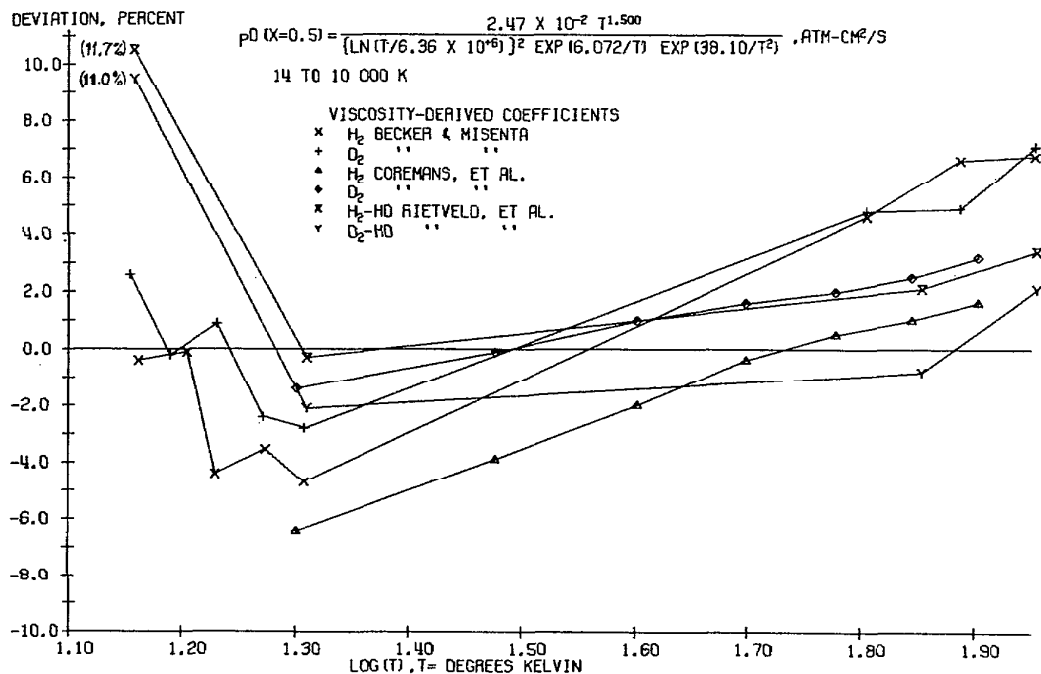


FIGURE 36. Deviations of diffusion coefficients from reference equation.
Hydrogen-Deuterium



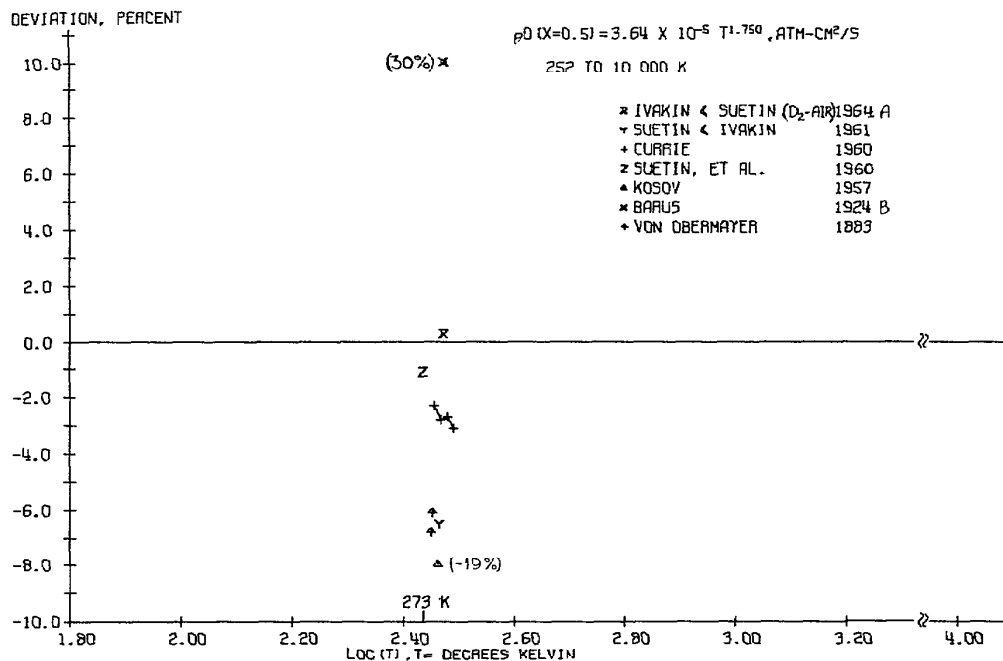


FIGURE 39. Deviations of diffusion coefficients from reference equation.

Hydrogen-Air

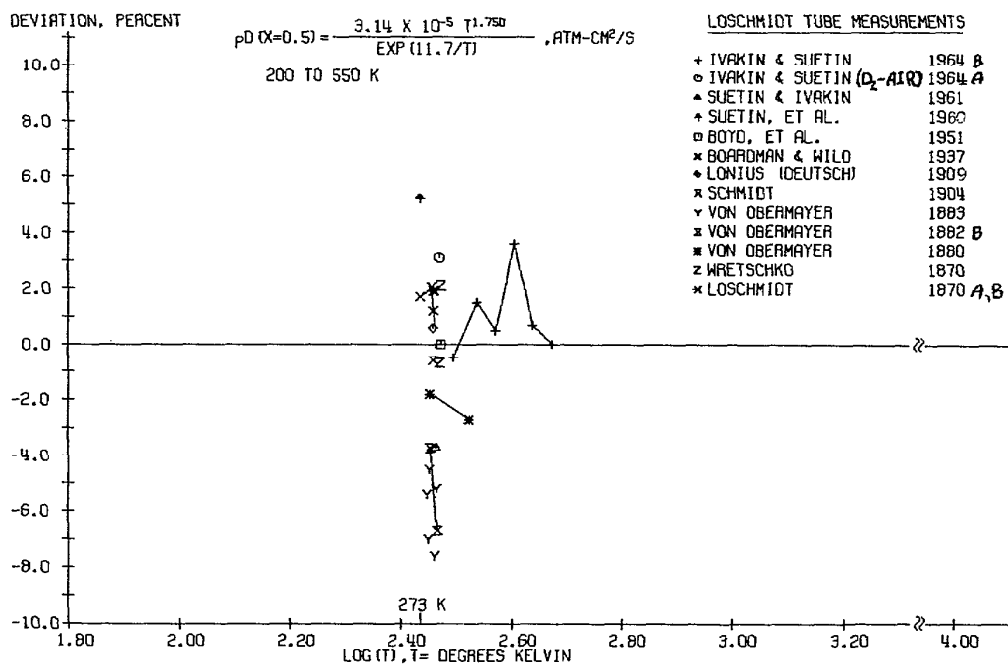


FIGURE 40. Deviations of diffusion coefficients from reference equation.

Hydrogen-Carbon dioxide

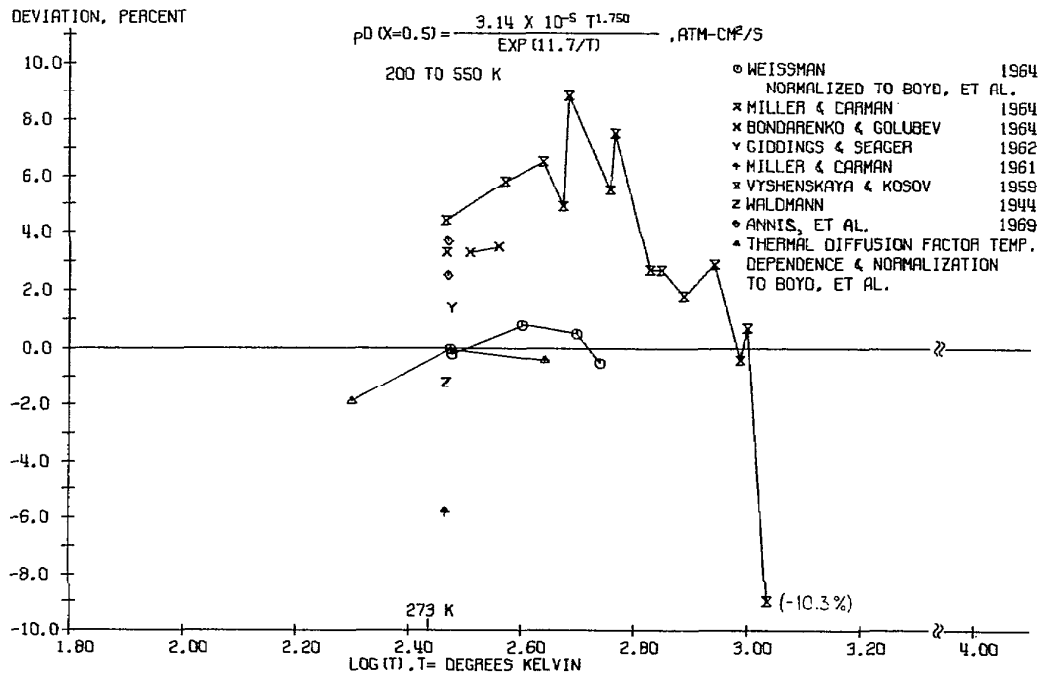


FIGURE 41. Deviations of diffusion coefficients from reference equation.

Hydrogen-Carbon dioxide

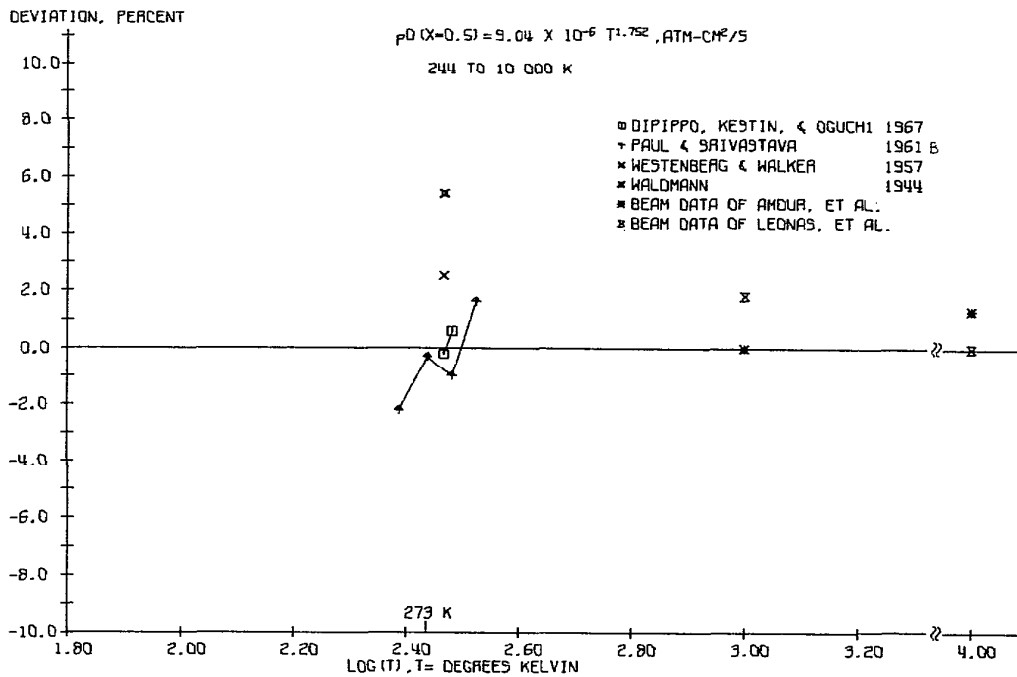


FIGURE 42. Deviations of diffusion coefficients from reference equation.

Nitrogen-Argon

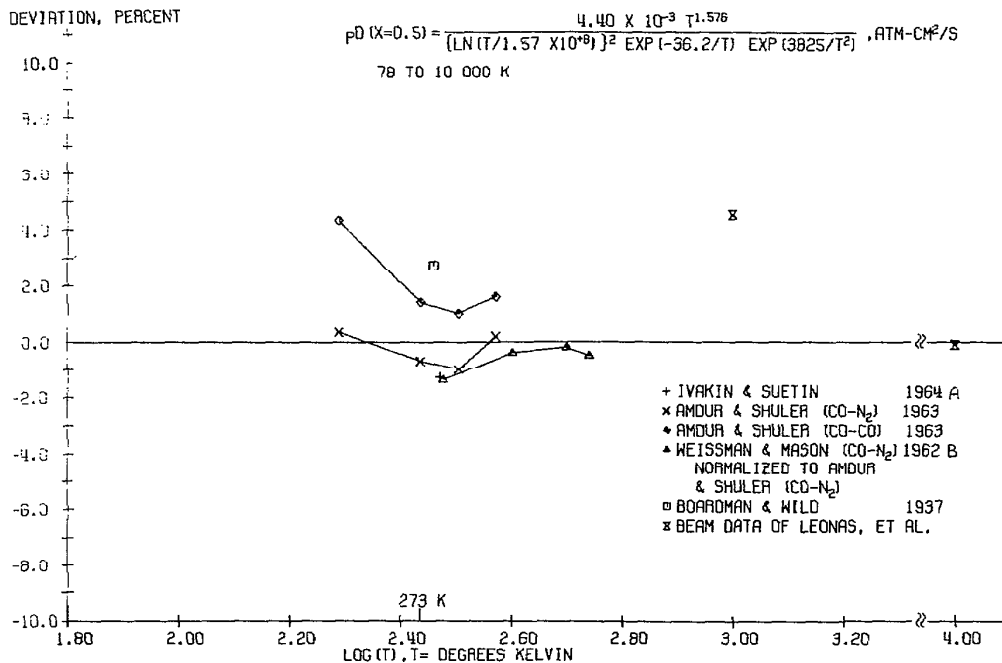


FIGURE 43. Deviations of diffusion coefficients from reference equation.

Carbon monoxide-Nitrogen

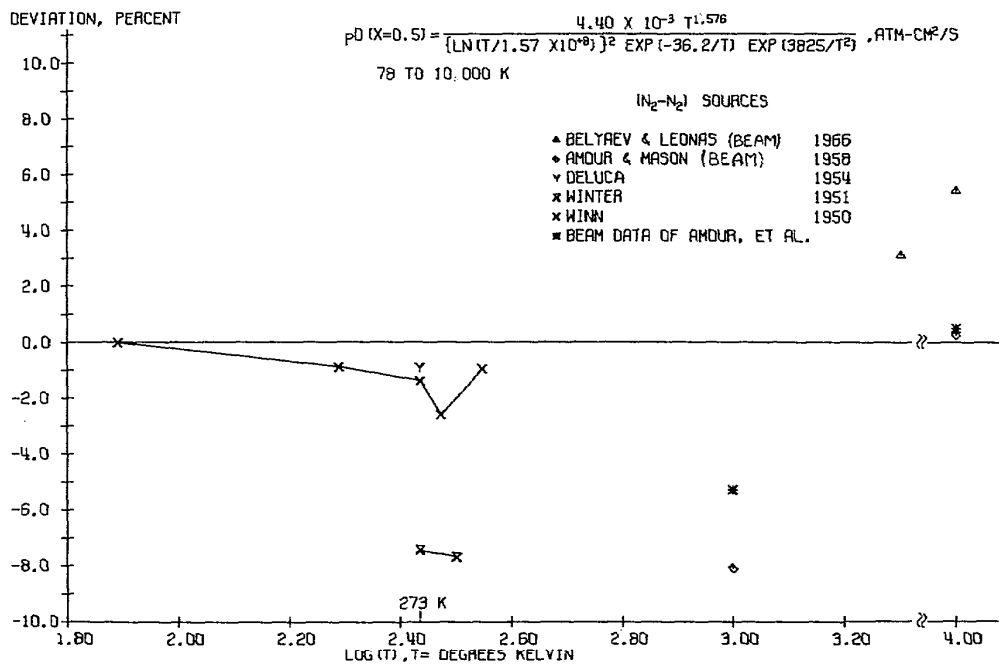


FIGURE 44. Deviations of diffusion coefficients from reference equation.

Carbon monoxide-Nitrogen

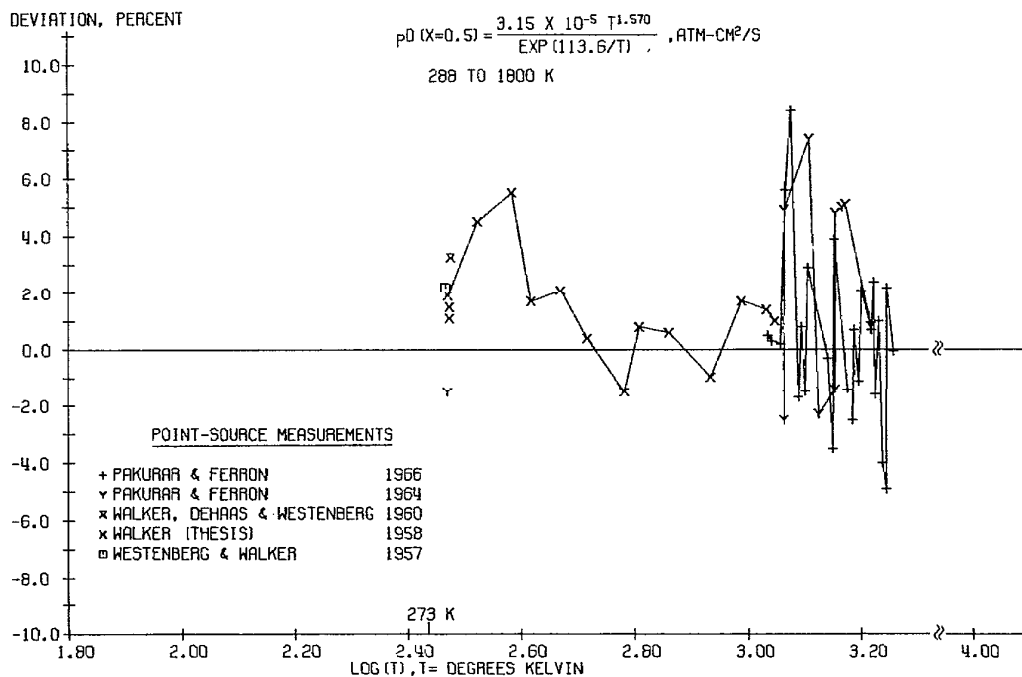


FIGURE 45. Deviations of diffusion coefficients from reference equation.

Nitrogen-Carbon dioxide

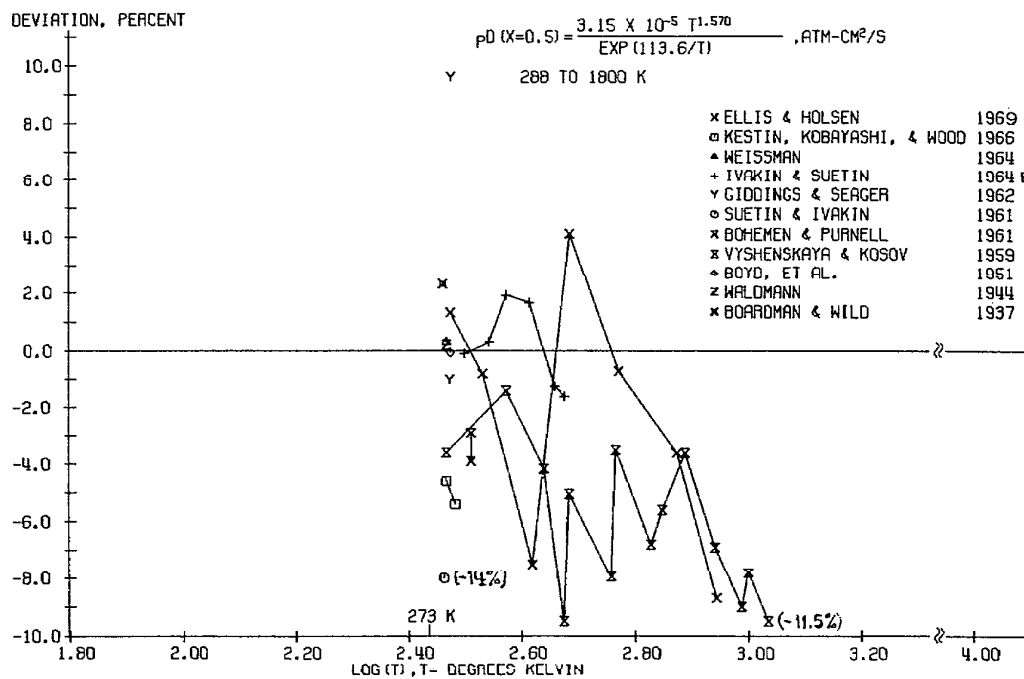


FIGURE 46. Deviations of diffusion coefficients from reference equation.

Nitrogen-Carbon dioxide

lack of measurements at low temperatures and asymmetry between the CO and N₂ molecules which may lead to small differences between the potentials.

He-O₂. This gas pair is a borderline system of Group II. He-O₂ was assigned to Group II on the strength of the two-bulb measurements by Paul and Srivastava (1961 a) and the consistency of its \mathcal{D}_{12} with those of He-N₂ and He-air.

He-air and H₂-air. These two gas pairs are grouped together because their reference equations have been calculated by application of Blanc's law, eq (2.1-7); that is, He-air was obtained from He-N₂ and He-O₂, and H₂-air from H₂-N₂ and H₂-O₂. The reliable determinations of \mathcal{D}_{12} have been by the closed-tube method; namely, for He-air the room temperature point from Fedorov et al. (1966) and for H₂-air a point from Ivakin and Suetin (1964 a). Each of these direct measurements is in excellent agreement with the specified reference equations. However, since Blanc's law introduces a small uncertainty into the results, the H₂-air system was not assigned to Group I along with H₂-N₂, but to Group II.

Each lower temperature limit for the reference equations of He-air and H₂-air has been set by data of He-O₂ (244 K) and H₂-O₂ (252 K), respectively. But the lower temperature limit for He-N₂ is 77 K, and for H₂-N₂ it is 65 K. In order to estimate \mathcal{D}_{12} for He-air at lower temperatures, if necessary, it is suggested that the ratio of \mathcal{D}_{12} for He-O₂ to He-N₂ at room temperature be taken as a constant, independent of temperature, and a similar procedure used for H₂-air. By applying this ratio the correlation range can be extended for He-O₂ and H₂-O₂, as well as He-air and H₂-air to the lower temperature limits for He-N₂ (77 K) and for H₂-N₂ (65 K).

He-CO₂. The few closed-tube measurements by Holsen and Strunk (1964) and by Ivakin and Suetin (1964 b), plus one two-bulb measurement by Annis et al. (1969), established He-CO₂ as a Group II gas pair.

H₂-He. This gas pair is in Group II primarily on the basis of reliable closed-tube measurements by Amdur and Malinauskas (1965), by Rumpel (1955), and by Bunde (1955).

H₂-Ne. This gas pair is a borderline Group II system because only one set of direct measurements of \mathcal{D}_{12} is available, obtained by the two-bulb method (Paul and Srivastava, 1961 c).

H₂-Ar. There are many independent determinations of \mathcal{D}_{12} for H₂-Ar, but with a number of significant discrepancies, see figures 32 and 33. The most reliable results are probably by Westenberg and Frazier (1962). The usually reliable closed-tube measurements by Ivakin and Suetin (1964 b) seem to give too steep a temperature dependence for \mathcal{D}_{12} . The only direct molecular-beam measurement for H₂-Ar gives a potential which is suspected to be too great (Colgate et al., 1969). Thus the molecular-beam potential for H₂-Ar was obtained by application of the combination rules, see table 21.

H₂-Kr. The gas pair H₂-Kr was placed in Group II on the basis of the measurements by Fedorov et al. (1966) and Annis et al. (1968). The temperature dependence of the thermal diffusion factor (Annis et al., 1968) was used to extend the correlation temperature range from room temperature down to 77 K. Near the temperature limits of the reference equation the data may not be as reliable as the Group II

uncertainty limits, thus H₂-Kr is considered a borderline system.

H₂-D₂. Most of the \mathcal{D}_{12} for H₂-D₂ have been calculated from viscosity measurements of the H₂ isotopes or mixtures thereof, see figures 36 and 37. At low temperatures the viscosity-derived \mathcal{D}_{12} are considered more reliable than the direct measurements. None of the direct measurements of \mathcal{D}_{12} were actually used for curve-fitting purposes. In the least-squares calculations the value of s of eq (4.3-1) turned out to be 1.4883 which was rounded to 1.500 to agree with the theoretical lower limit for the rigid-sphere model, and the remaining correlation constants were determined on the basis that $s=1.500$.

H₂-CO₂. The most reliable results for this gas pair are closed-tube measurements by Loschmidt (1870 b), Boyd et al. (1951), and Ivakin and Suetin (1964 b). The results by Vyshenskaya and Kosov (1959) extend to 1083 K, but these have not been used to establish the upper temperature limit of the reference equation because the data were not considered sufficiently reliable, see figure 41. The viscosity-derived \mathcal{D}_{12} reported by Weissman (1964) were normalized to the datum of Boyd et al. (1951) because the points seemed systematically high by about 5 percent.

N₂-Ar. The N₂-Ar gas pair is in Group II on the basis of the measurements of Paul and Srivastava (1961 b) and one point-source measurement of Westenberg and Walker (1957); however, these results cover only a small temperature range, so that this is a borderline system.

N₂-CO. On the basis of two-bulb measurements by Winn (1950) and closed-tube measurements by Amdur and Shuler (1963), the gas pair N₂-CO was placed in Group II. Since N₂ and CO are isosteric molecules, the N₂-N₂, CO-CO, and N₂-CO data could all be used for \mathcal{D}_{12} . Determinations of \mathcal{D}_{12} from molecular-beam measurements have been published by Belyaev and Leonas (1966) and Amdur and Mason (1958) for N₂-N₂, and are in excellent agreement with the present calculations, see figure 44. The viscosity-derived \mathcal{D}_{12} reported by Weissman and Mason (1962 b) seemed systematically low, thus the published results were normalized to the datum at 319 K of Amdur and Shuler (1963).

N₂-CO₂. This gas pair is a Group II system on the basis of the measurements by Boyd et al. (1951), Walker and Westenberg (1958 a), Walker et al. (1960), and Pakurar and Ferron (1964, 1966). The results of Vyshenskaya and Kosov (1959) drop well below the recommended \mathcal{D}_{12} at high temperatures; a similar trend was evident for H₂-CO₂.

c. Group III (Deviation Plots, Figs. 47 to 75)

Weights and Potentials. The equimolar values of \mathcal{D}_{12} and their weights used in the least-squares calculations are presented in table 22. The potential functions obtained from molecular-beam measurements are summarized in table 23. Since there were no molecular-beam measurements available for systems with air or CO₂, no potentials are listed for the systems air-(Ar, CH₄, CO, SF₆) and CO₂-(Ar, CO, O₂, air, N₂O, SF₆). The potentials for most of the Group III gas pairs required the application of the combination rules. At present, Leonas et al. have not performed beam experiments with CH₄ and SF₆, but both of these gases have been used in experi-

ments by Amdur et al. Thus there are no independent confirmations of the potentials of gas pairs containing CH_4 or SF_6 . The directly measured potential for He-CH_4 was used to derive a potential for $\text{H}_2\text{-CH}_4$; but the $\text{CH}_4\text{-Ar}$ potential was used for the heavier gas pairs with methane; $\text{CH}_4\text{-(N}_2, \text{O}_2, \text{SF}_6)$. If the He-CH_4 potential had been used for $\text{CH}_4\text{-N}_2$, etc., inconsistent results would have been obtained. Apparently the small helium atom "sees" some of the structure of the CH_4 molecule, and the He-CH_4 potential is not generally suitable for combination-rule calculations based on the assumption of

spherically symmetric potentials (Mason and Amdur, 1964). The combination-rule potentials by Amdur et al. for $\text{H}_2\text{-O}_2$, $\text{CH}_4\text{-O}_2$, CO-Kr , CO-O_2 , and $\text{N}_2\text{-O}_2$ were obtained from two different apparatuses. Such results from "mixed" apparatus are reliable over a smaller temperature range than potentials obtained from the same equipment.

Special Comments. For the thirty-two gas pairs of Group III the special comments are as follows.

Ar- CH_4 . The closed-tube measurements by Arnold and Toor (1967) were considered sufficiently reliable to establish Ar- CH_4 as a Group III system.

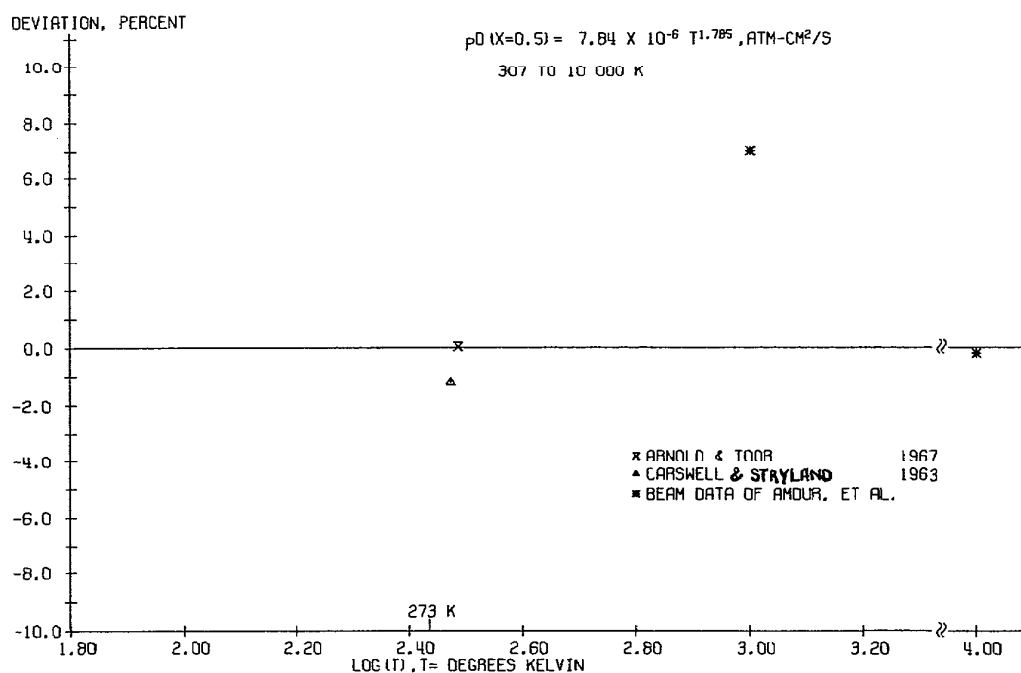


FIGURE 47. Deviations of diffusion coefficients from reference equation.

Methane-Argon

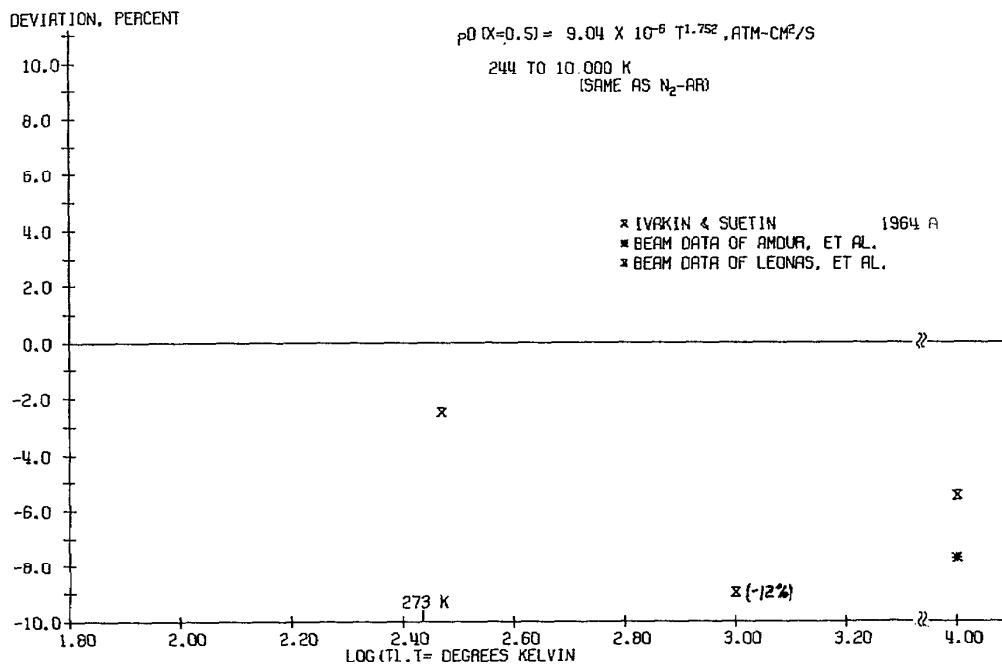


FIGURE 48. Deviations of diffusion coefficients from reference equation.
Carbon monoxide-Argon

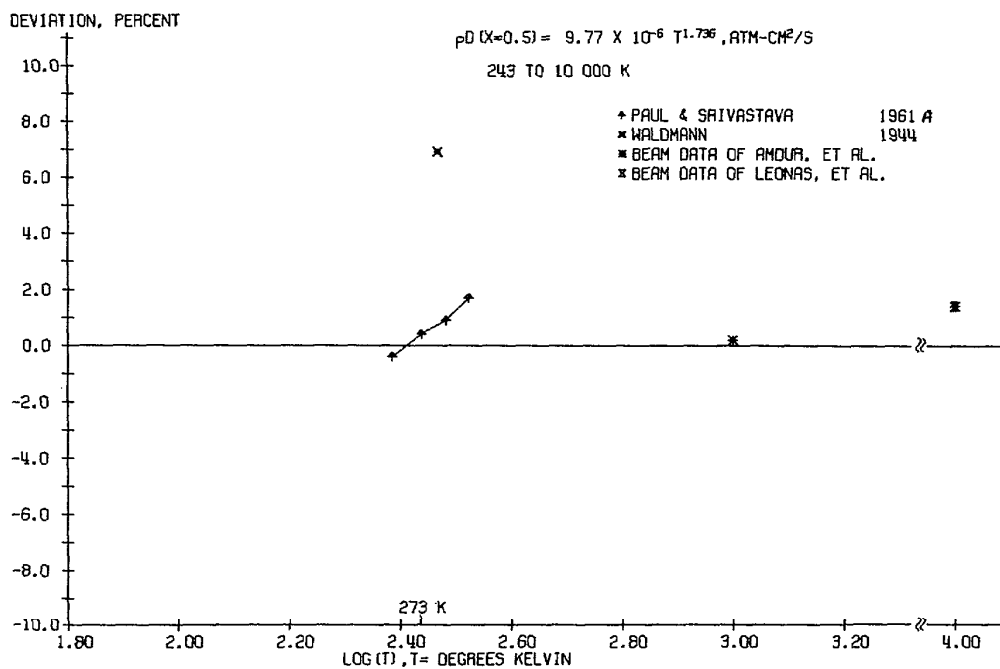


FIGURE 49. Deviations of diffusion coefficients from reference equation.
Oxygen-Argon

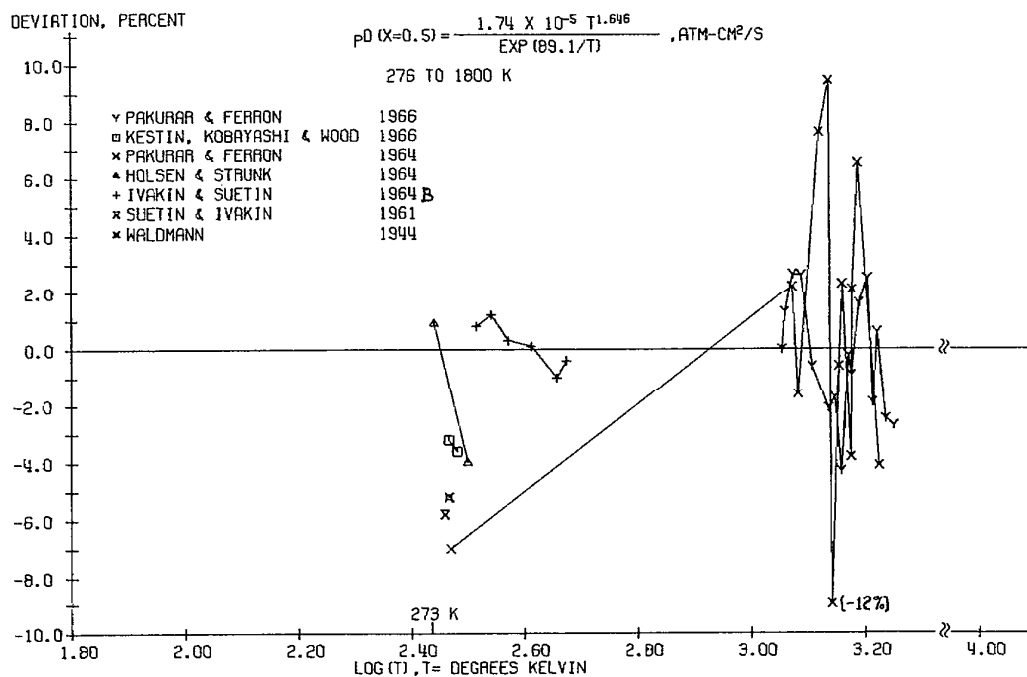


FIGURE 50. Deviations of diffusion coefficients from reference equation.

Argon-Carbon dioxide

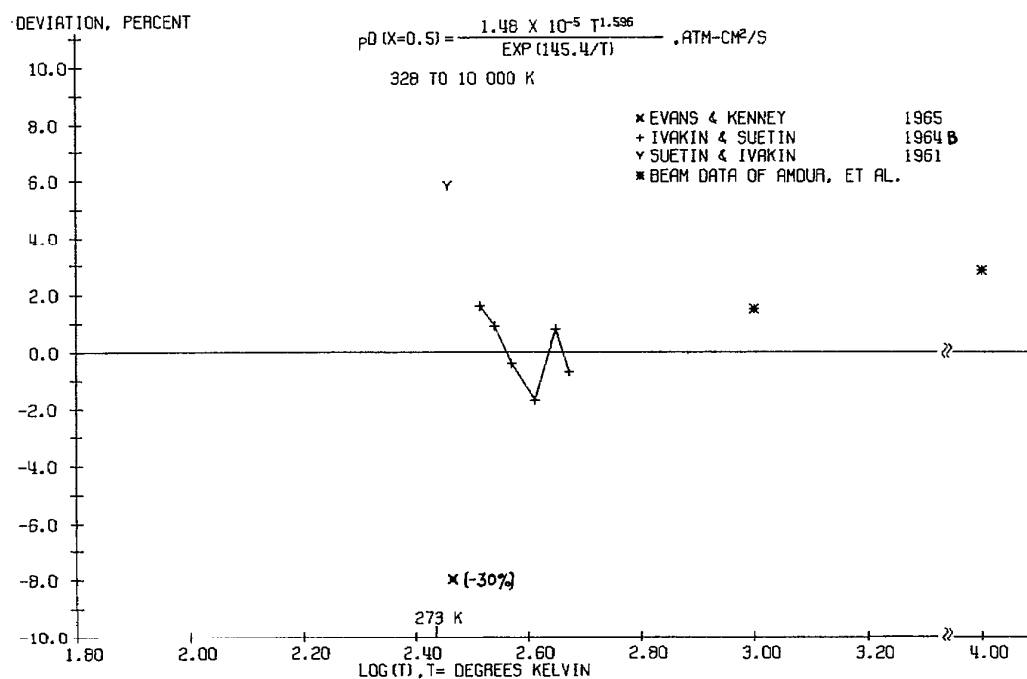


FIGURE 51. Deviations of diffusion coefficients from reference equation.

Argon-SF₆

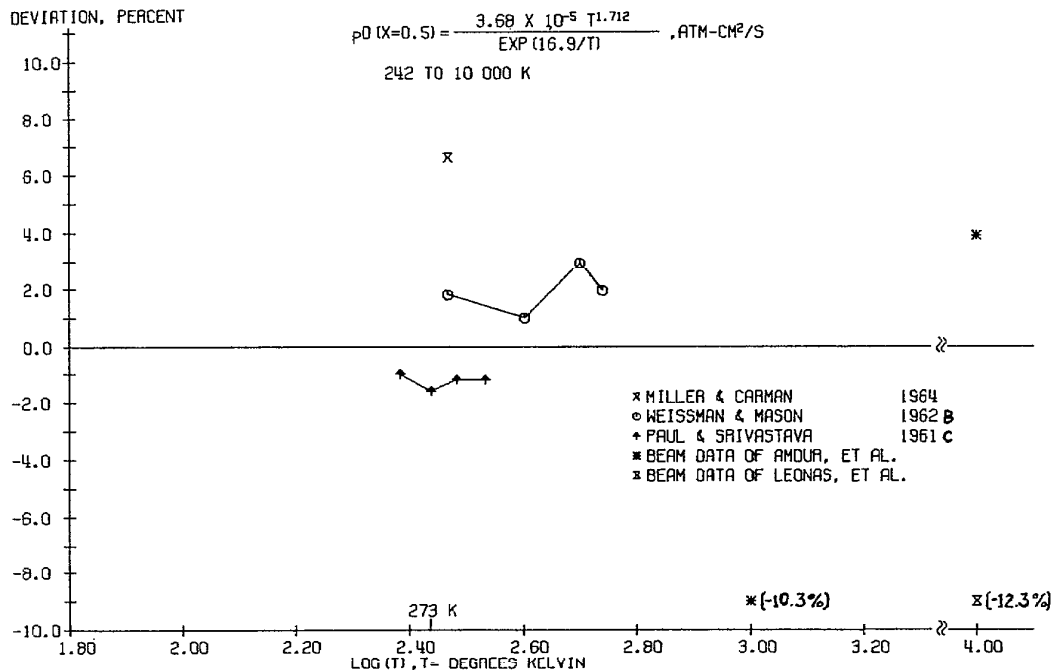


FIGURE 52. Deviations of diffusion coefficients from reference equation.

Hydrogen-Xenon

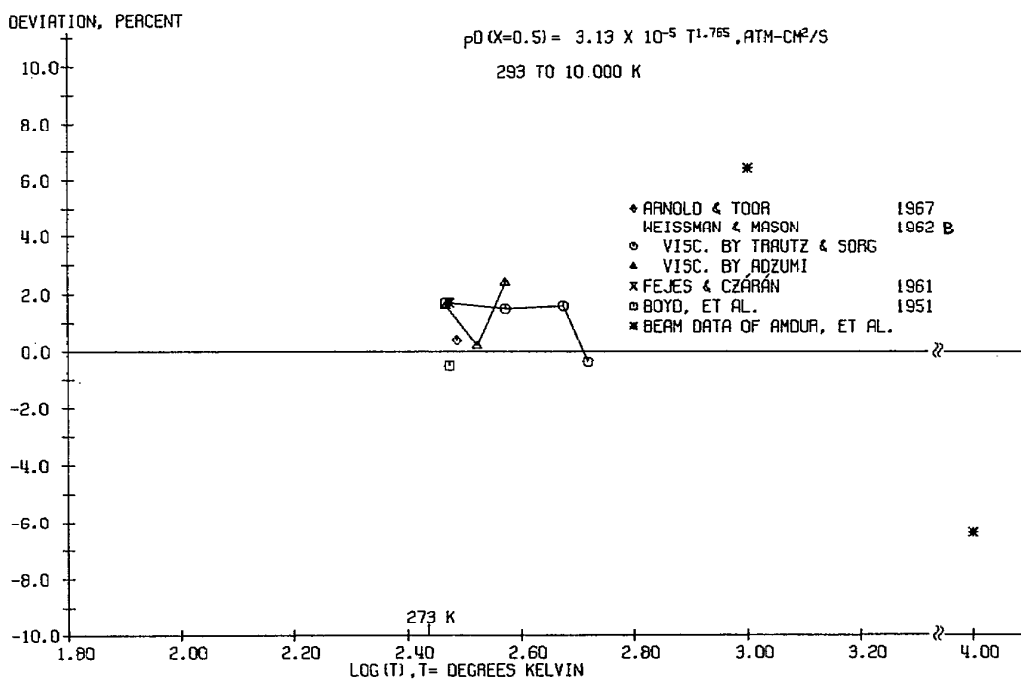


FIGURE 53. Deviations of diffusion coefficients from reference equation.

Hydrogen-Methane

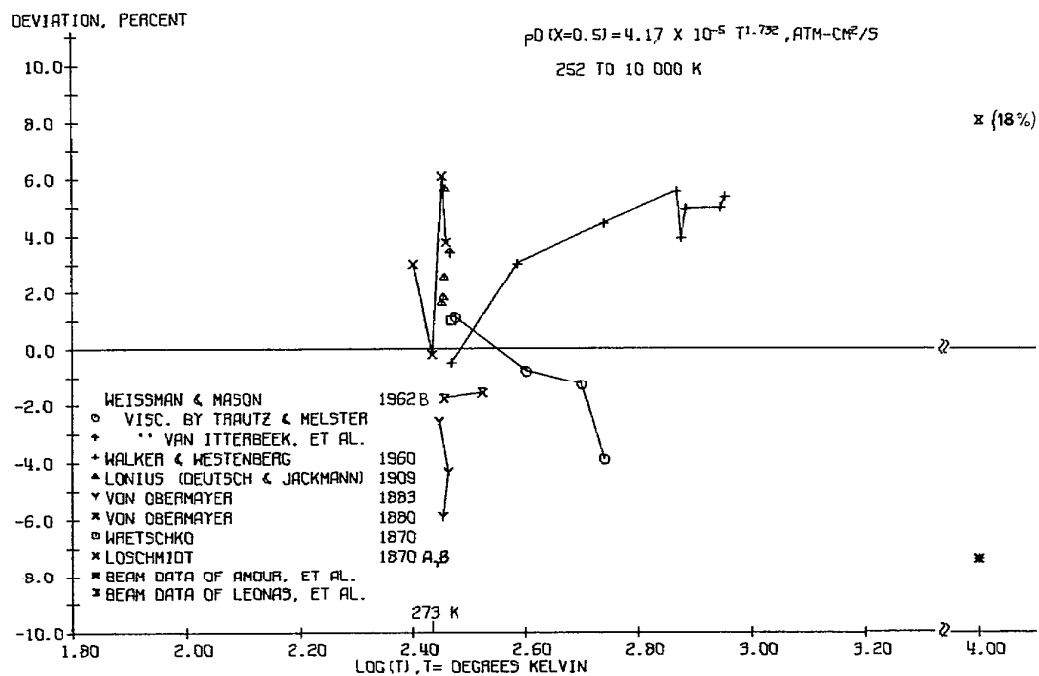


FIGURE 54. Deviations of diffusion coefficients from reference equation.

Hydrogen-Oxygen

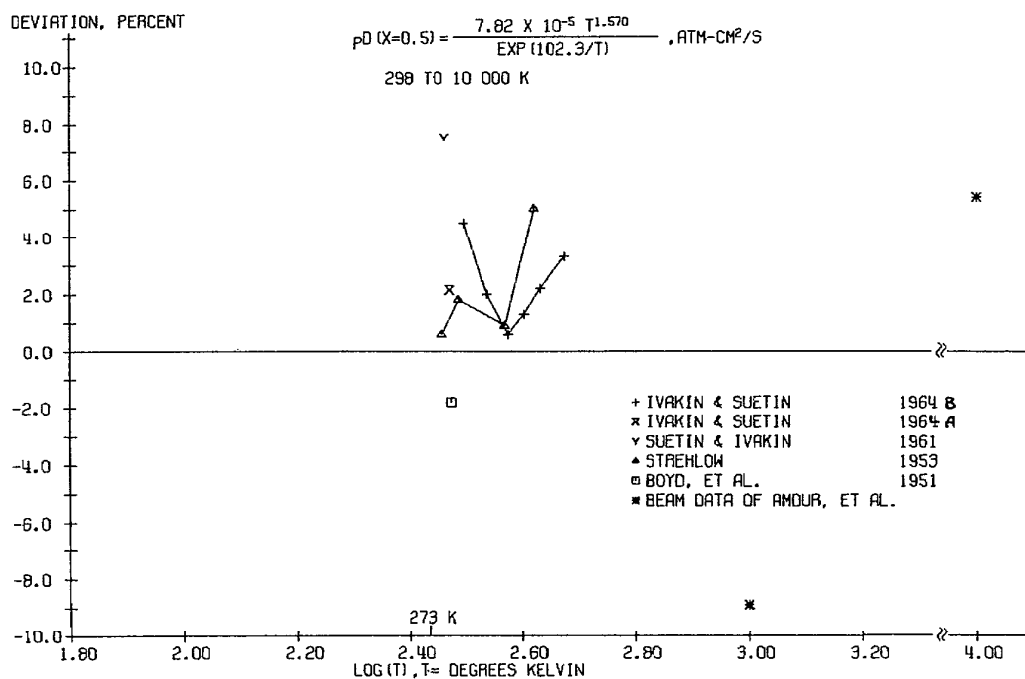


FIGURE 55. Deviations of diffusion coefficients from reference equation.

Hydrogen-SF₆

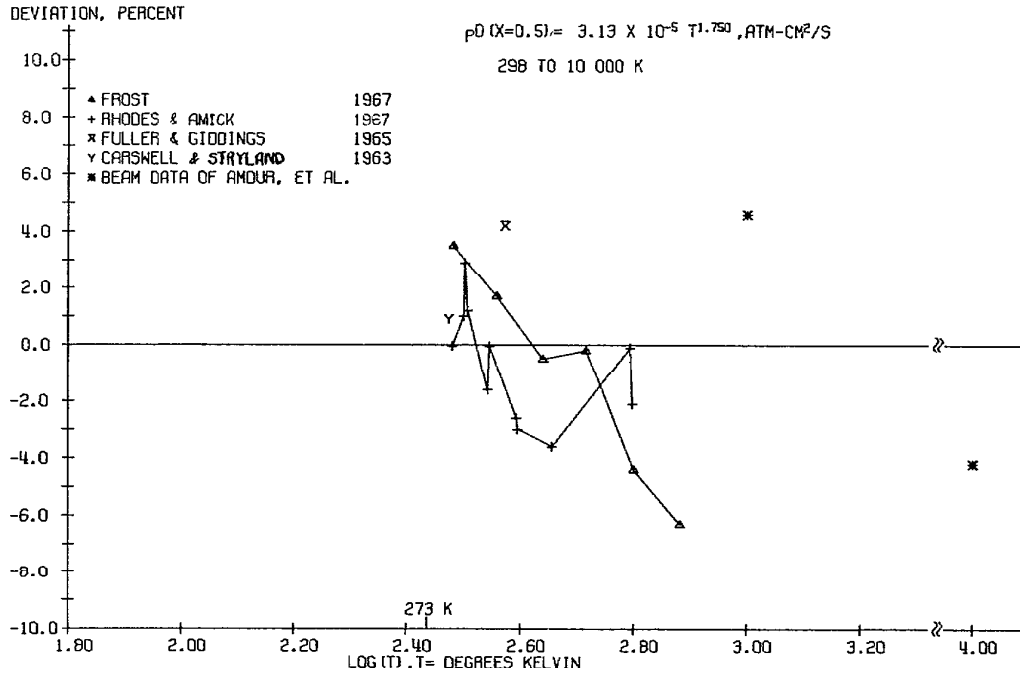


FIGURE 56. Deviations of diffusion coefficients from reference equation.

Helium - Methane

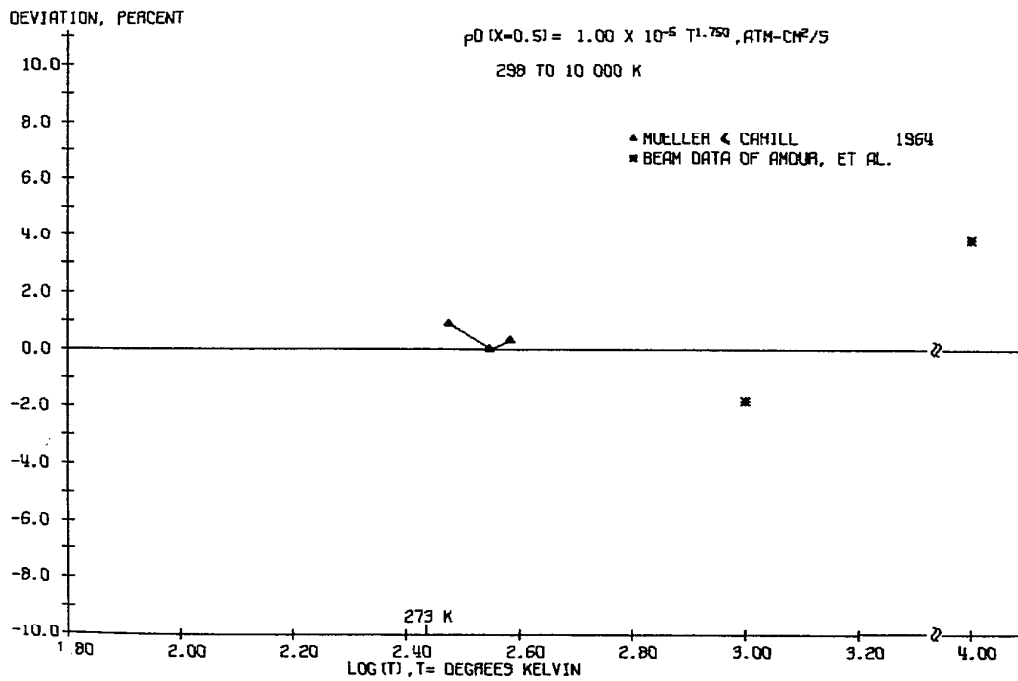


FIGURE 57. Deviations of diffusion coefficients from reference equation.

Methane - Nitrogen

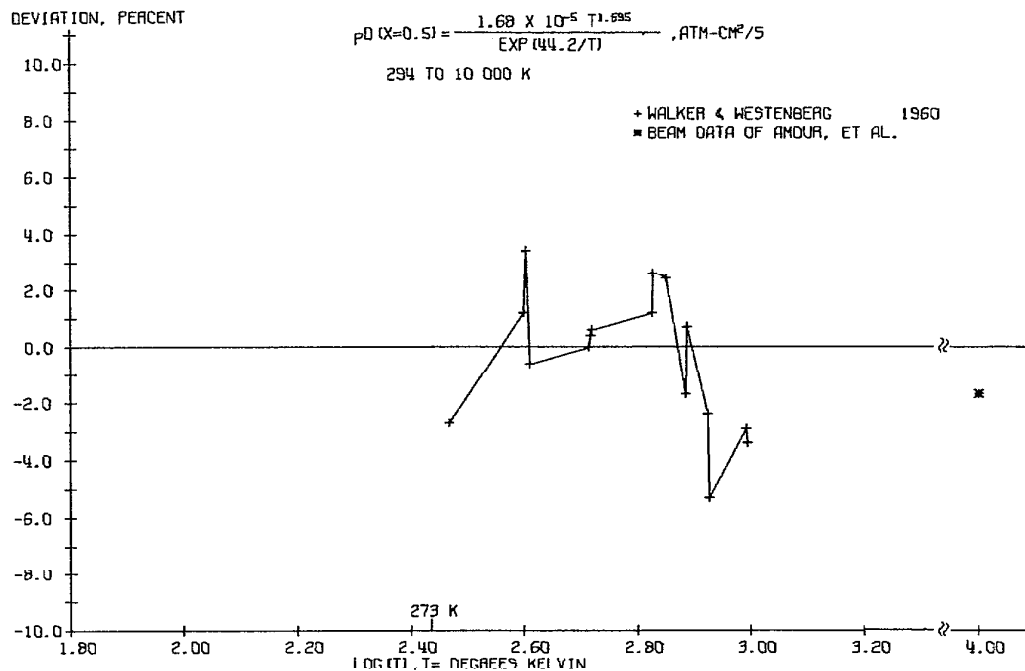


FIGURE 58. Deviations of diffusion coefficients from reference equation.

Methane-Oxygen

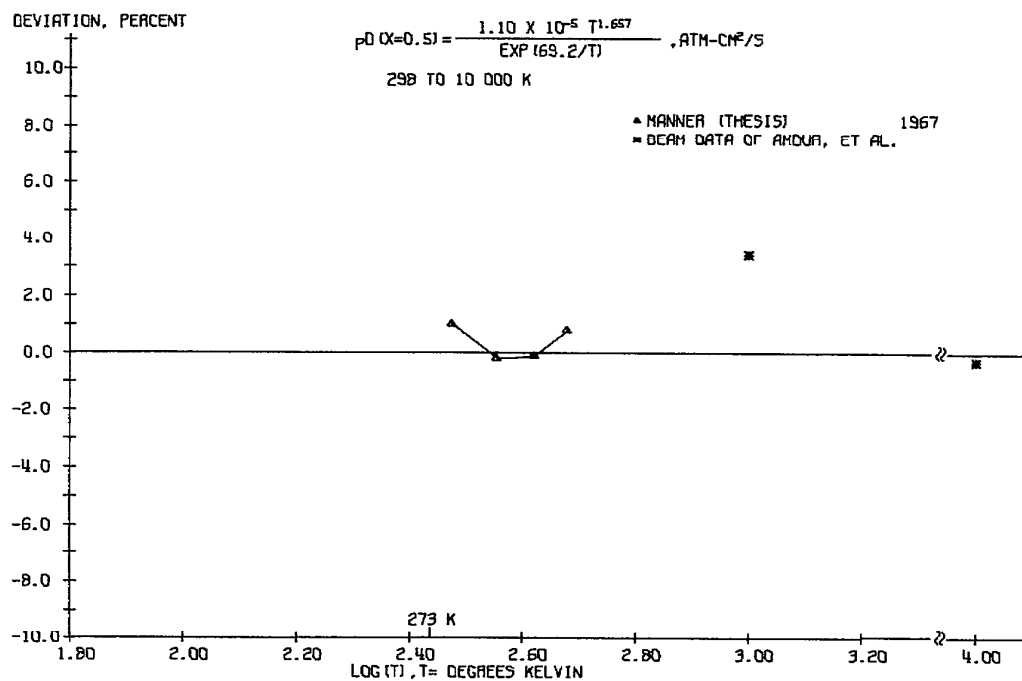


FIGURE 59. Deviations of diffusion coefficients from reference equation.

Methane-SF₆

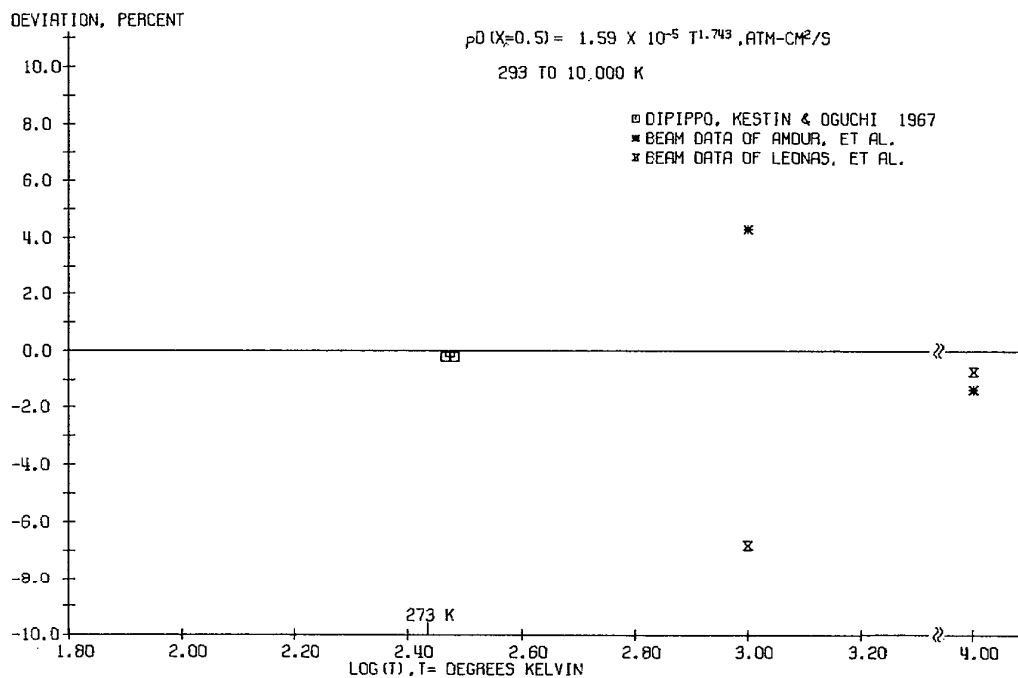


FIGURE 60. Deviations of diffusion coefficients from reference equation.
Neon-Nitrogen

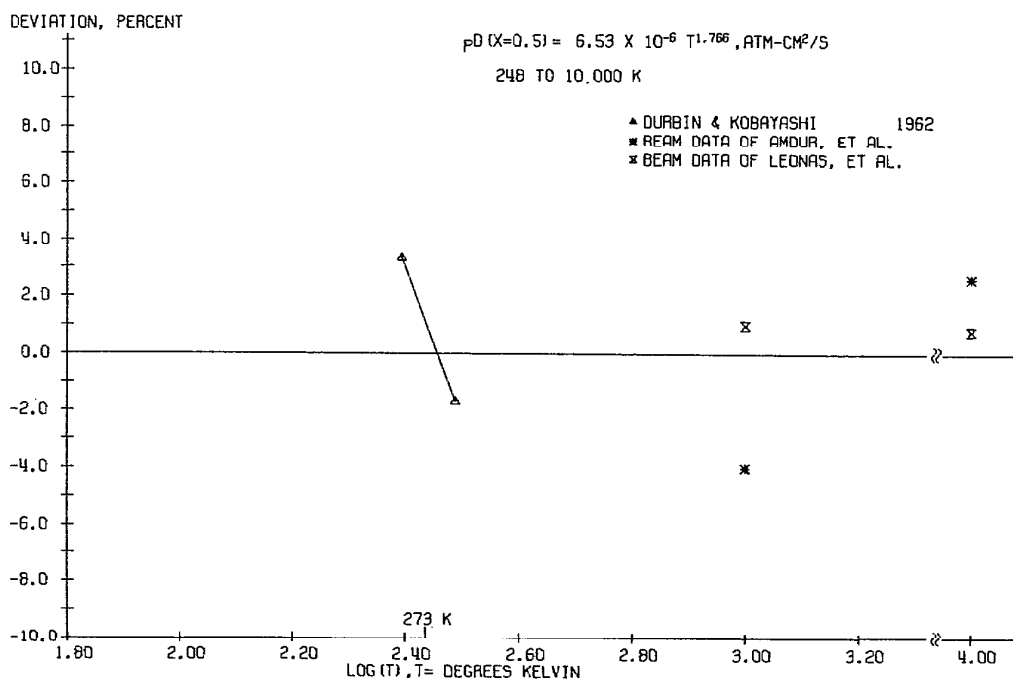


FIGURE 61. Deviations of diffusion coefficients from reference equation.
Nitrogen-Krypton

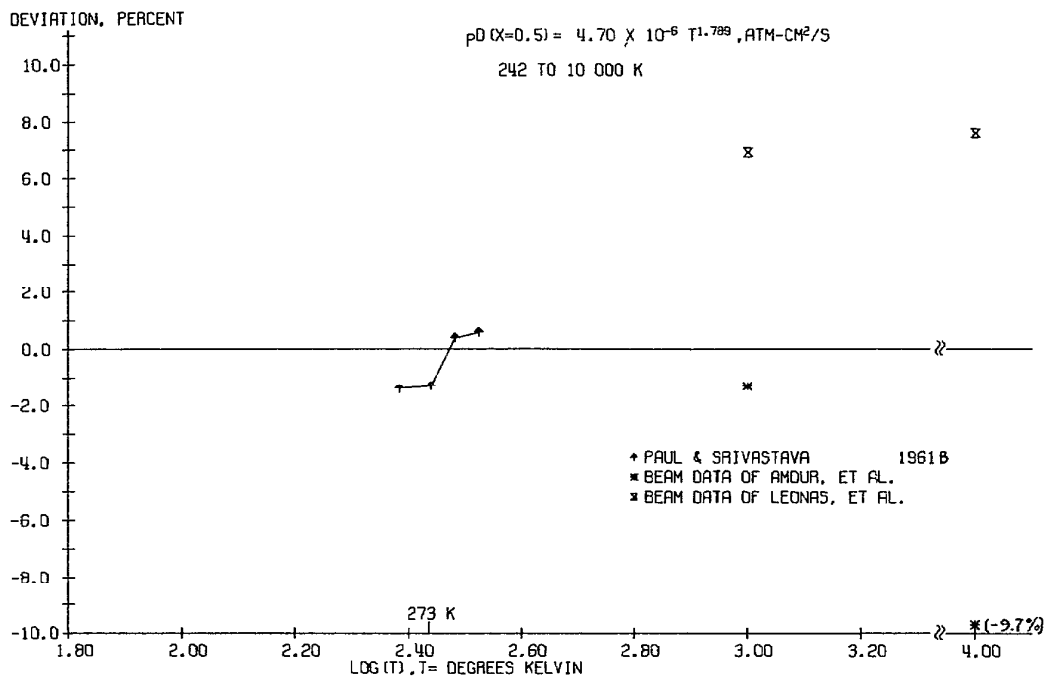


FIGURE 62. Deviations of diffusion coefficients from reference equation.

Nitrogen-Xenon

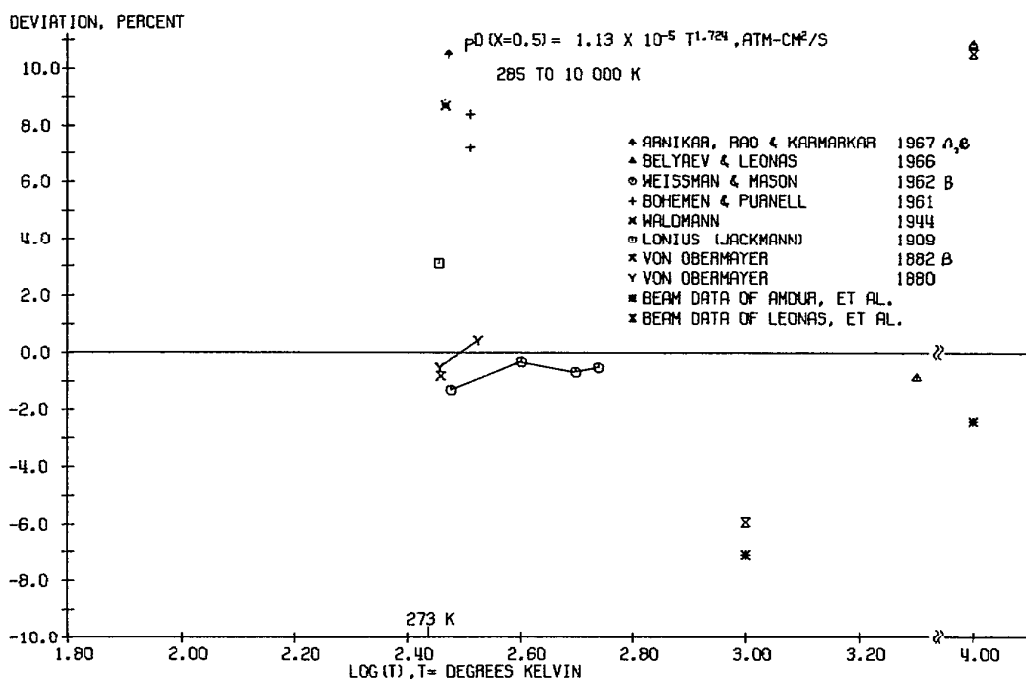


FIGURE 63. Deviations of diffusion coefficients from reference equation.

Nitrogen-Oxygen

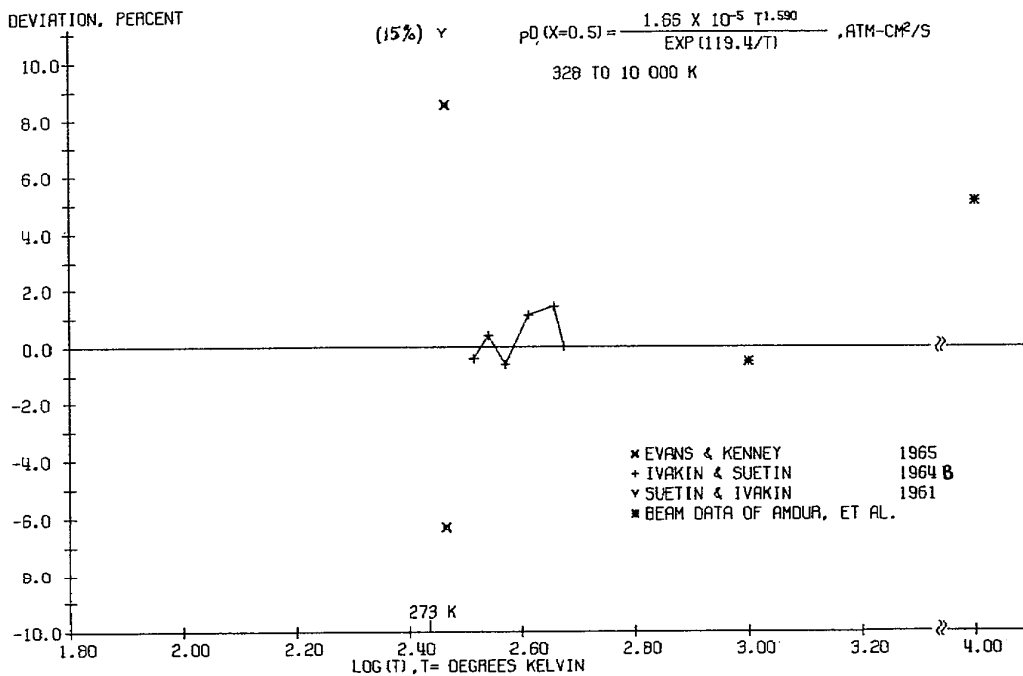


FIGURE 64. Deviations of diffusion coefficients from reference equation.
Nitrogen-SF₆

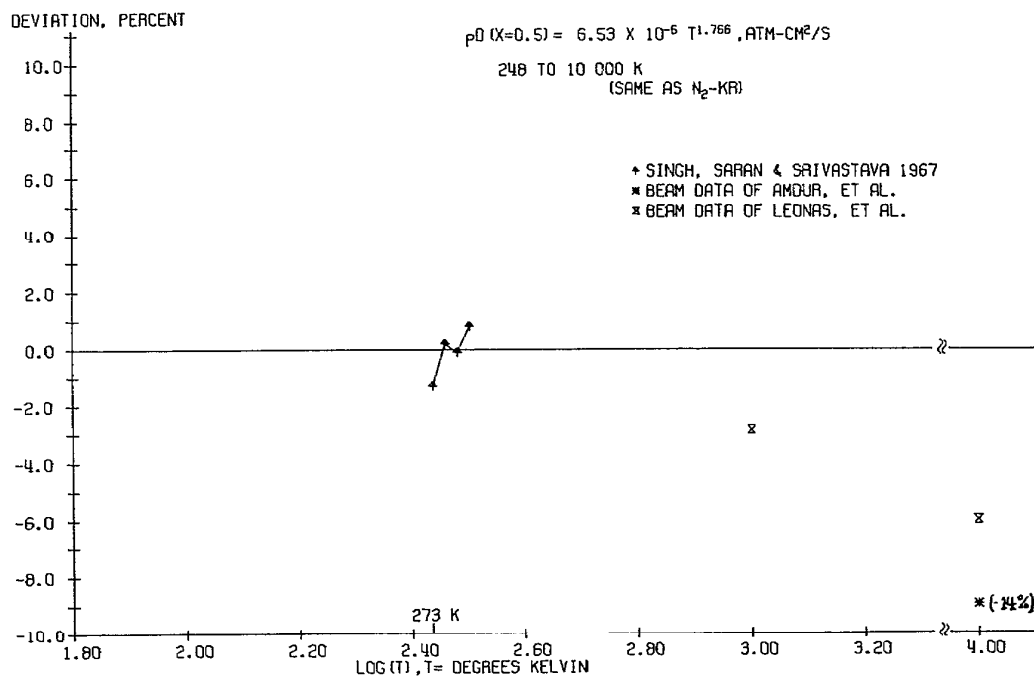


FIGURE 65. Deviations of diffusion coefficients from reference equation.
Carbon monoxide-Krypton

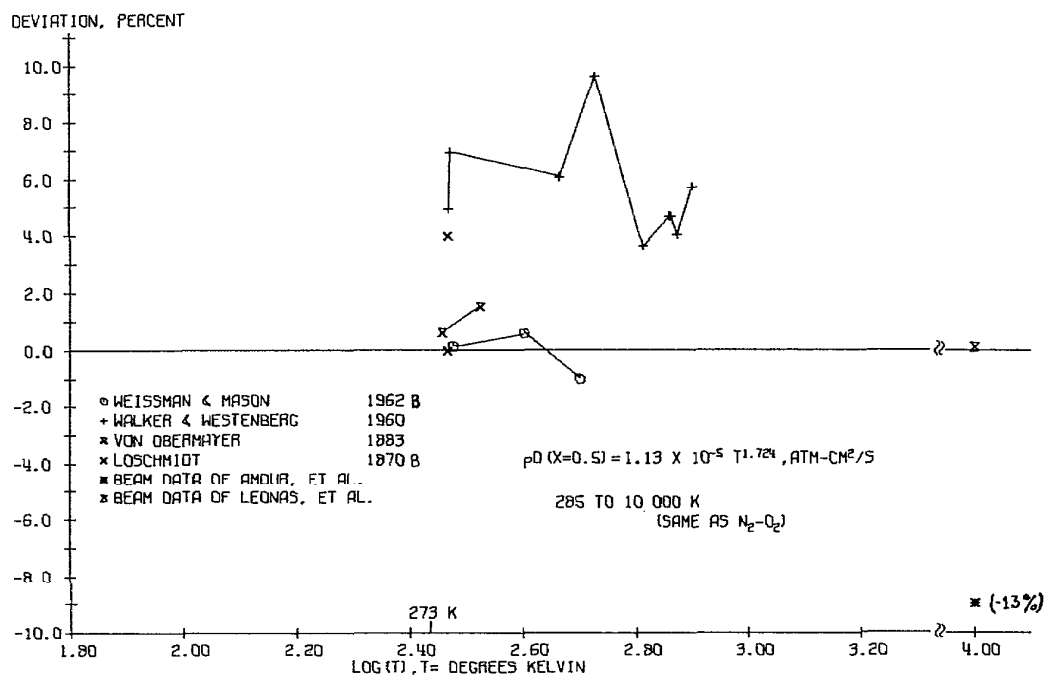


FIGURE 66. Deviations of diffusion coefficients from reference equation.

Carbon monoxide-Oxygen

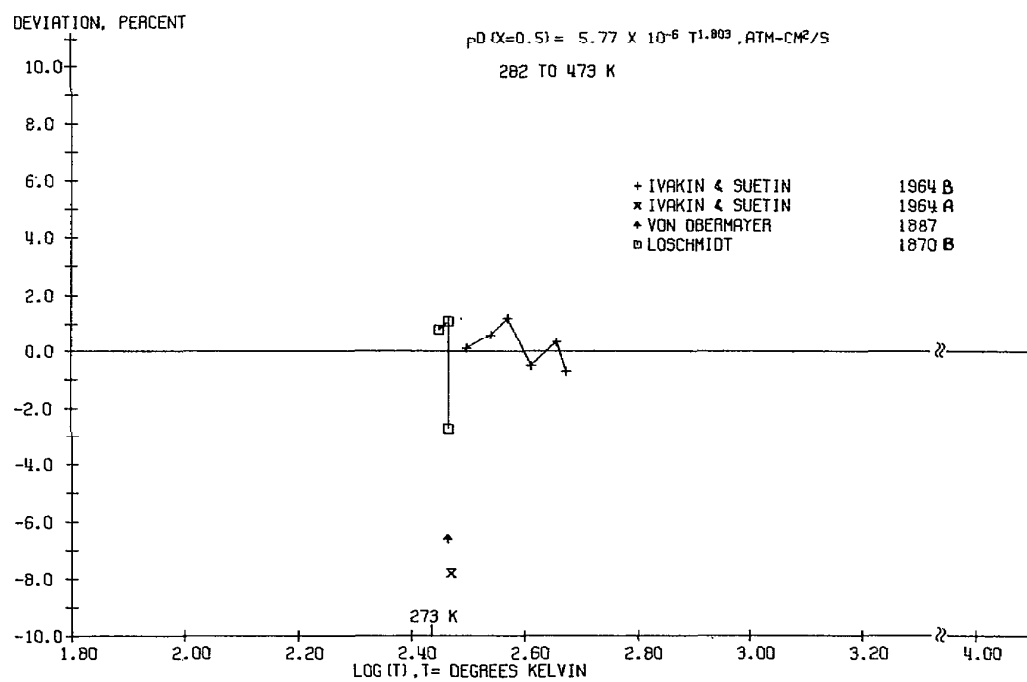


FIGURE 67. Deviations of diffusion coefficients from reference equation.

Carbon monoxide-Carbon dioxide

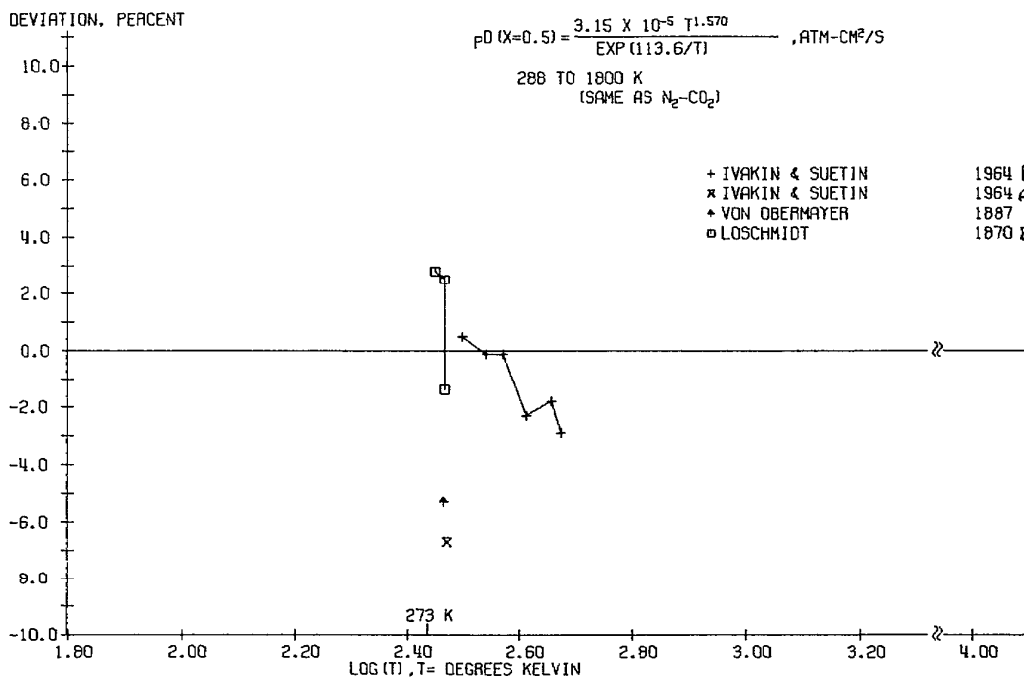


FIGURE 68. Deviations of diffusion coefficients from reference equation.
Carbon monoxide—Carbon dioxide

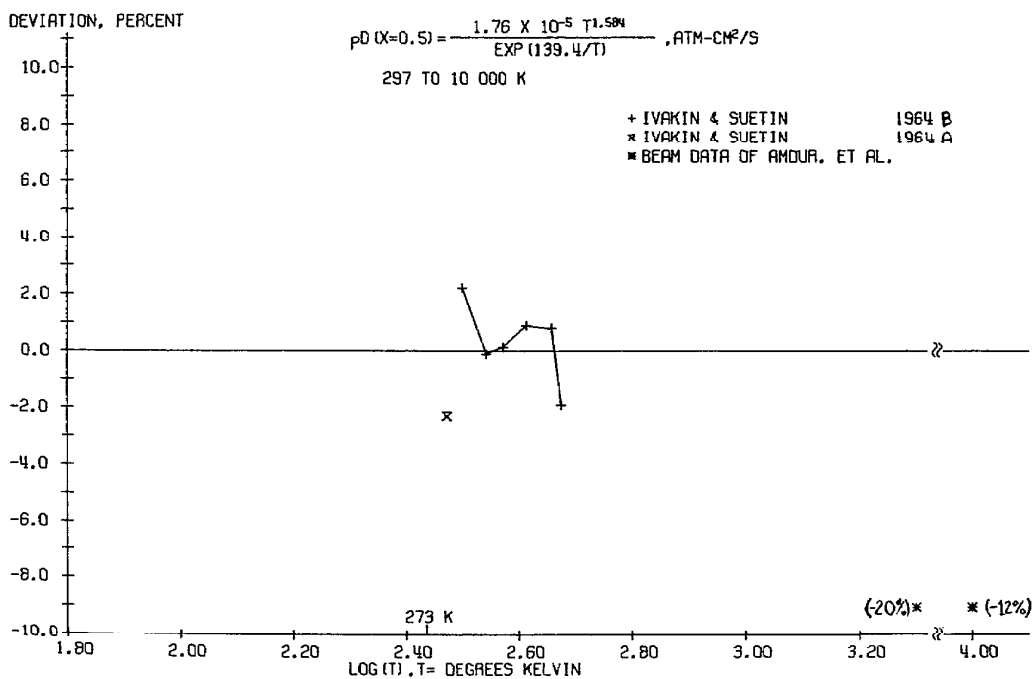


FIGURE 69. Deviations of diffusion coefficients from reference equation.
Carbon monoxide—SF₆

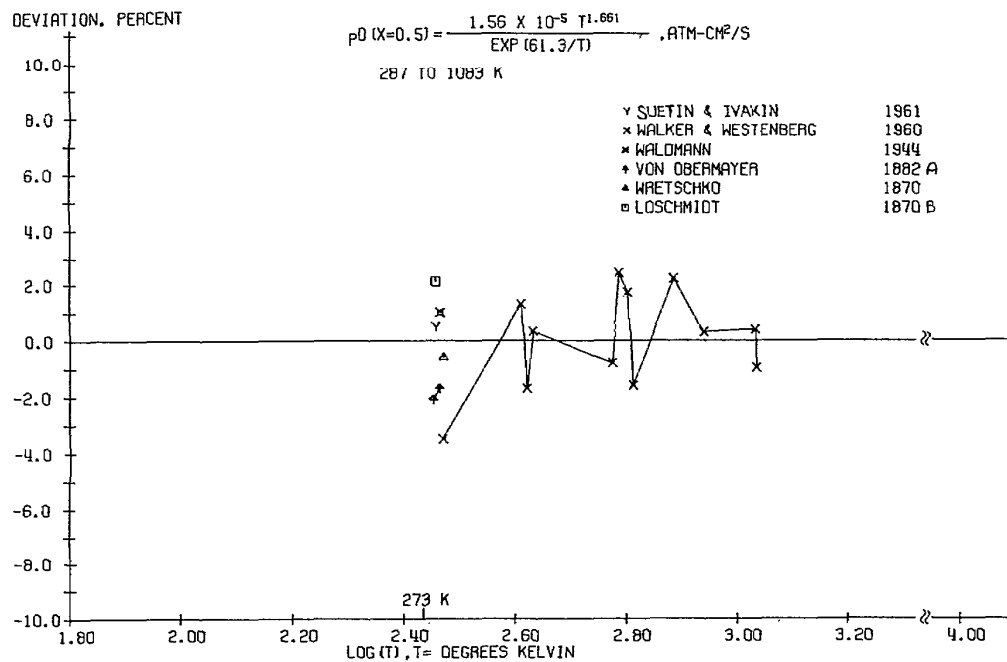


FIGURE 70. Deviations of diffusion coefficients from reference equation.

Oxygen-Carbon dioxide

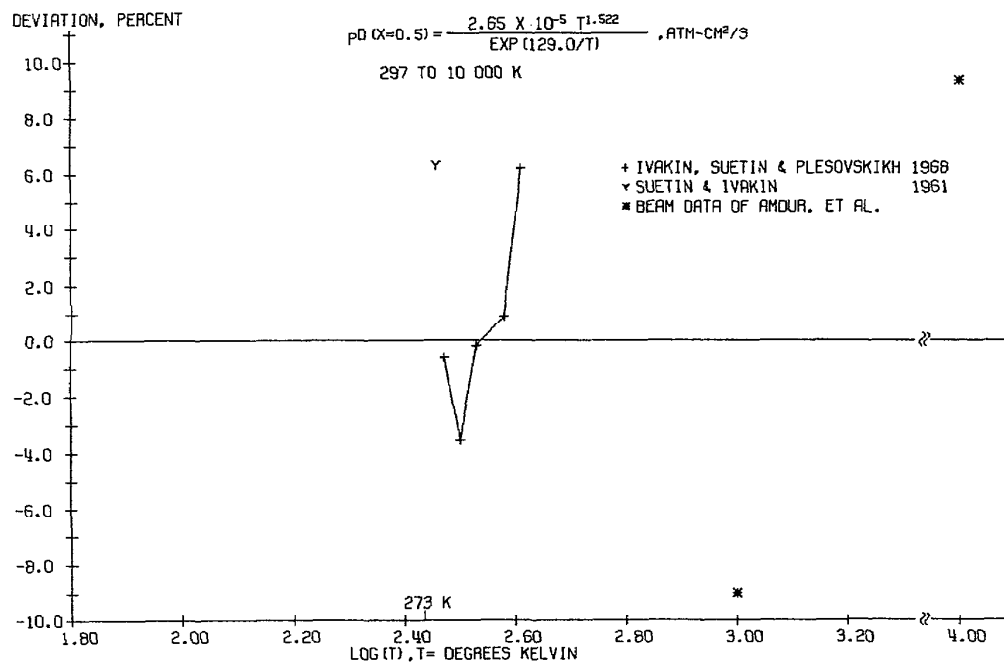


FIGURE 71. Deviations of diffusion coefficients from reference equation.

Oxygen-SF₆

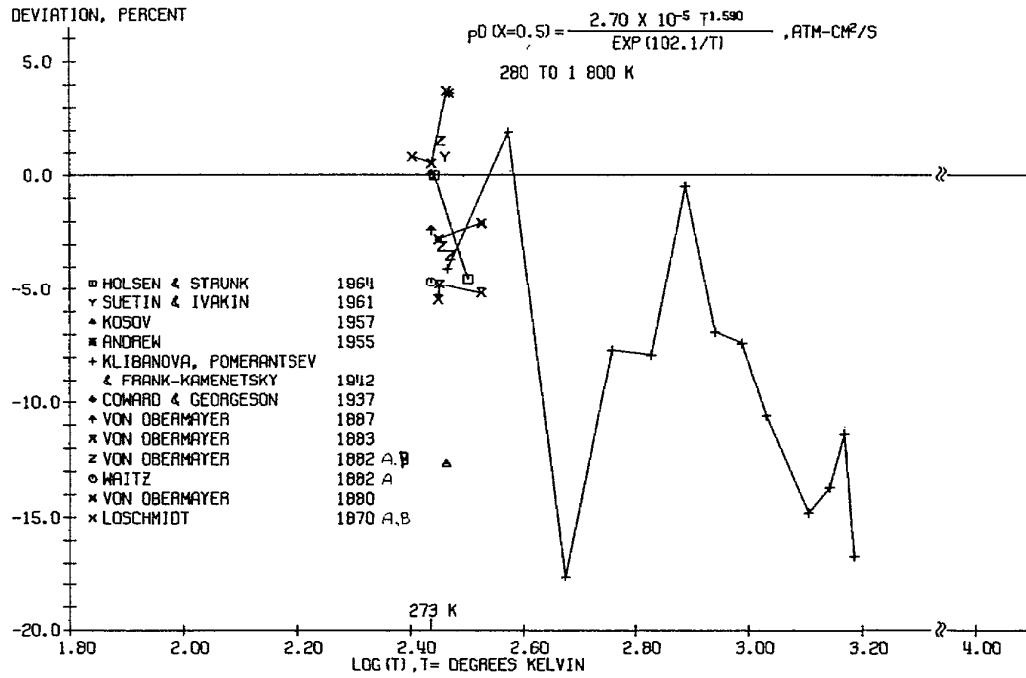


FIGURE 72. Deviations of diffusion coefficients from reference equation.

Air-Carbon dioxide

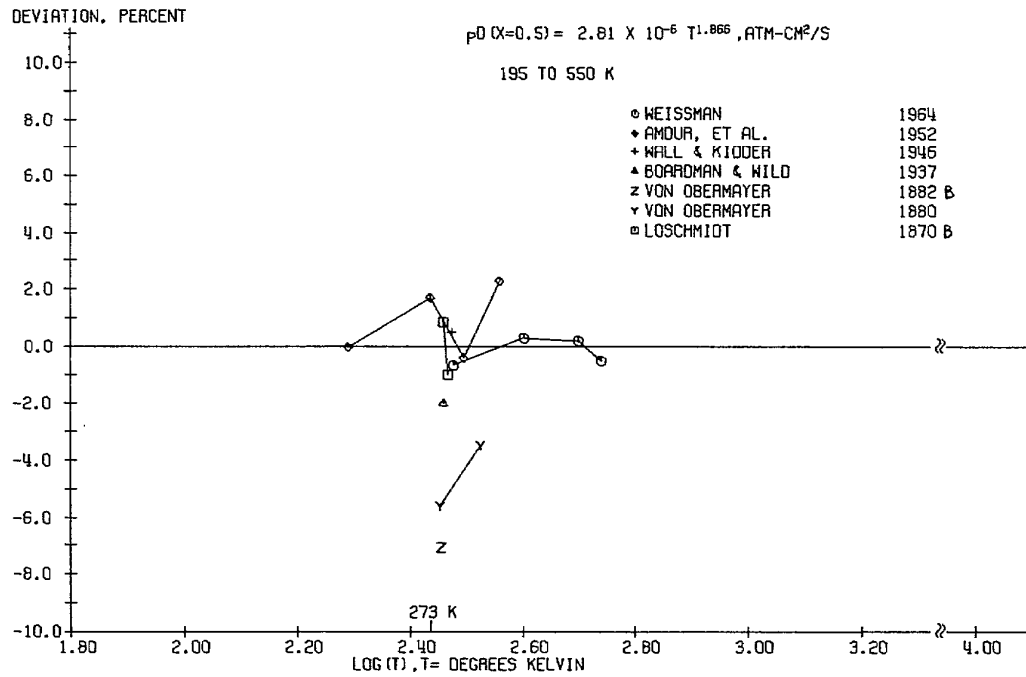


FIGURE 73. Deviations of diffusion coefficients from reference equation.

Carbon dioxide-N₂O

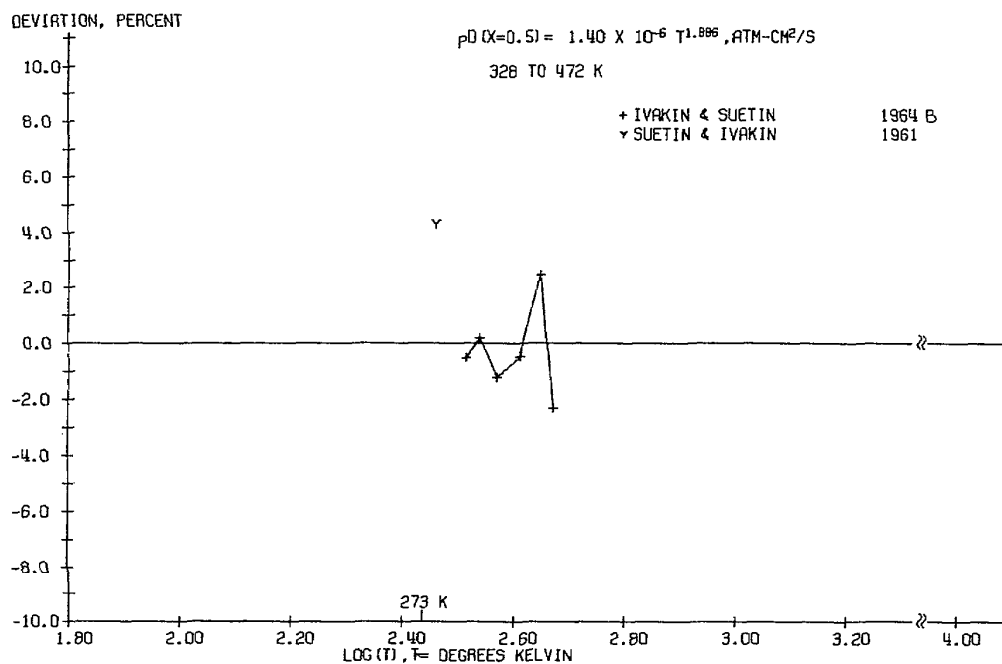


FIGURE 74. Deviations of diffusion coefficients from reference equation.

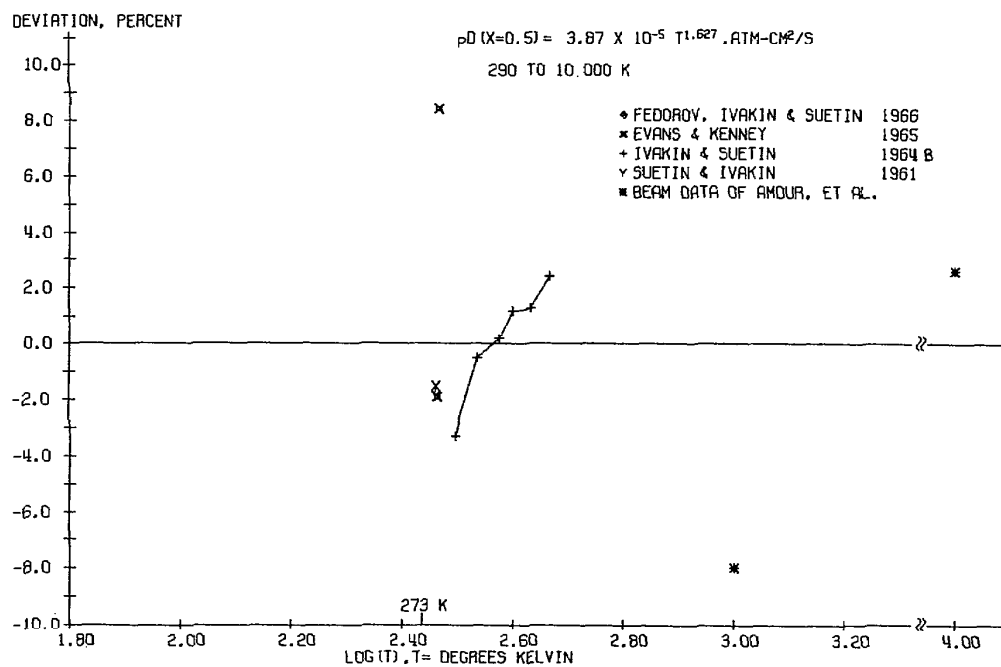
Carbon dioxide-SF₆

FIGURE 75. Deviations of diffusion coefficients from reference equation.

Helium-SF₆

TABLE 22. Diffusion coefficients and weights for curve-fitting, Group III^a

System	T, K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note	System	T, K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note
Ar-CH ₄	307.15	-0.6655	b	CH ₄ -SF ₆	297.58	-0.9566	o
	8660	1.9232	c		357.92	-.8119	o
Ar-CO	-	-	d		418.19	-.6872	o
Ar-O ₂	316	-0.670	*		477.87	-.5784	o
	3160	1.066	*		2045	0.5159	i
Ar-air	282	-0.7520	e		5000	1.0212	i
	355	-.5784	e	N ₂ -Ne	293.15	-.04989	p
	447	-.4029	e		6090	1.7973	i
	1000	.2095	e	N ₂ -Kr	316	-.0770	*
	10 000	1.9576	e		5620	1.438	*
Ar-CO ₂	276.2	-.08775	f	N ₂ -Xe	316	-.0855	*
	317.2	-.7820	f		7940	1.650	*
	328.0	-.7328	g	N ₂ -O ₂	316	-.0638	*
	348.0	-.6819	g		3160	1.086	*
	373.0	-.6289	g	N ₂ -SF ₆	328.0	-.0393	g
	410.0	-.5528	g		348.0	-.8861	g
	455.0	-.04737	g		373.0	-.8327	g
	473.0	-.4401	g		410.0	-.7471	g
	1100	.2122	h		455.0	-.6615	g
	1800	.5752	h		473.0	-.6364	g
Ar-SF ₆	328.0	-1.0000	g		1000	-.0640	i
	348.0	-.09508	g		3960	.9294	i
	373.0	-.8962	g	CO-Kr	-	-	d
	410.0	-.8210	g	CO-O ₂	-	-	d
	447.0	-.7375	g	CO-air	282	-.7077	e
	472.0	-.6981	g		355	-.5376	e
	1000	-.0985	i		447	-.3686	e
	4640	1.0086	i		1000	.2279	e
H ₂ -Xe	242.2	-.03872	j		10 000	1.9727	e
	274.2	-.2941	j	CO-CO ₂	281.6 _s	-.08187	q
	303.9	-.2132	j		293.3 _s	-.7854	q
	341.2	-.1244	j		293.1 _s	-.8027	q
	293.2	-.2277	k		315.4	-.7328	g
	400.0	.0065	k		348.0	-.6536	g
	500.0	.1847	k		373.0	-.5969	g
	550.0	.2529	k		410.0	-.5302	g
	2320	1.2989	i		455.0	-.4449	g
	8290	2.2856	i		473.0	-.4191	g
H ₂ -CH ₄	316	-0.092	*	CO-SF ₆	296.0	-1.0521	g
	10 000	2.556	*		315.4	-.09788	g
H ₂ -O ₂	316	-0.050	*		348.0	-.9031	g
	3160	1.682	*		373.4	-.8416	g
H ₂ -SF ₆	298.15	-.03788	l		410.0	-.7595	g
	286.2	-.4023	m		455.0	-.6737	g
	306.9	-.3391	m		473.0	-.6536	g
	370.8	-.1891	m		1000	-.0640	i
	418.0	-.0768	m		3960	0.9294	i
	313.0	-.3006	g	O ₂ -CO ₂	286.9 _s	-.8069	q
	344.4	-.2441	g		287.1 _s	-.8097	q
	376.0	-.1791	g		287.1 _s	-.8041	q
	401.0	-.1249	g		296.5 _s	-.7932	r
	429.0	-.0680	g		287.8	-.8125	s
	473.0	.0128	g		296	-.8069	s
	1320	.7185	i		409	-.5287	n
	7460	1.9782	i		419	-.5229	n
CH ₄ -He	316	-0.130	*		430	-.4935	n
	3160	1.620	*		596	-.2457	n
CH ₄ -N ₂	316	-0.625	*		612	-.2111	n
	10 000	2.000	*		635	-.1858	n
CH ₄ -O ₂	293.6 _s	-.06676	n		649	-.1838	n
	395	-.4168	n		768	-.0434	n
	402	-.3936	n		770	-.0297	n
	408	-.3990	n		864	0.0302	n
	517	-.2122	n		867	.0546	n
	521	-.2048	n		874	.0492	n
	534	-.1993	n		1080	.2084	n
	668	-.0101	n		1081	.2098	n
	669	-.0031	n		1083	.2049	n
	707	.0378	n	O ₂ -SF ₆	297	-1.0044	t
	708	.0418	n		317	-.09626	t
	768	.0831	n		340	-.8894	t
	771	.0973	n		379	-.7959	t
	840	.1523	n		408	-.7144	t
	842	.1467	n		2930	.658	i
	845	.1399	n		6310	1.212	i
	3550	1.250	*	CO ₂ -air	282	-.08300	e
	10 000	2.000	*		355	-.6387	e
CH ₄ -air	282	-0.7077	e		501	-.3636	e
	355	-.5317	e		708	-.0992	e
	447	-.3551	e		1000	.1575	e
	1000	0.2577	e		1590	.4921	e
	10 000	2.0009	e				

TABLE 22. Diffusion coefficients and weights for curve-fitting, Group III^a — Continued

System	T, K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note	System	T, K	$\log_{10}[\mathcal{D}_{12}(x=1/2)]$	Note
CO ₂ -N ₂ O	287.9 ₅	-.9586	q	CO ₂ -SF ₆	328.0	-1.1113	g
	287.9 ₅	-.9582	q		348.0	-1.0595	g
	293.1 ₅	-.9520	q		373.0	-1.0088	g
	288.1 ₅	-.9706	u		410.0	-0.9281	g
	298.1 ₅	-.9318	v		447.0	-.8446	g
	194.8	-1.2790	w		472.0	-.8210	g
	273.2	-0.9974	w	SF ₆ -He	316	-.345	*
	312.8	-.9052	w		10 000	2.095	*
	312.8	-.8887	w	SF ₆ -air	300	-1.0097	e
	362.6	-.7657	w		500	-0.5901	e
	300.0	-.9318	x		700	-.3298	e
	400.0	-.6946	x		1000	-.0630	e
	500.0	-.5143	x		10 000	1.5599	e
550.0	-.4401	x					

* Selected value, see explanation in first part of section 5.4.

^a All listed values of \mathcal{D}_{12} are weighted one; except in H₂-SF₆ for which the datum by Boyd et al. (1951) at 298.15 K is weighted ten.^b Arnold and Toor (1967).^c Calculated from molecular-beam potential by Mason and Amdur (1964), see table 23.^d Reference equation of N₂(gas) is suitable because of isosteric molecules.^e Calculated from reference equations for N₂(gas) and O₂(gas) according to Blanc's law, eq (2.1-7).^f Holsen and Strunk (1964).^g Ivakin and Suetin (1964 b).^h Pakurar and Ferron (1966) and Ferron (1967).ⁱ Calculated from molecular-beam potential by Amdur et al., see table 23.^j Paul and Srivastava (1961 c).^k Weissman and Mason (1962 b).^l Boyd et al. (1951); weight of datum ten for least-squares calculations.^m Sirehlow (1953).ⁿ Walker and Westenberg (1960, 1966).^o Manner (1967).^p DiPippo et al. (1967).^q Loschmidt (1870 b).^r Wretschko (1870).^s Suetin and Ivakin (1961).^t Ivakin et al. (1968).^u Boardman and Wild (1937).^v Wall and Kidder (1946).^w Amdur et al. (1952).^x Weissman (1964).Table 23. Molecular-beam potentials, $\phi(r) = K/r^s$, for Group III^{a, b}

System	Potential			Source			Reference
	K, eV(Å) ^s	s	Range, Å	system	K, eV(Å) ^s	s	
Ar-CH ₄	936	7.85	2.31 -2.66	Direct measurement			Mason and Amdur (1964).
Ar-CO	551	6.99	2.09 -2.68	Direct measurement			Jordan et al. (1970).
	580	7.14 ₅	2.28 ₅ -3.03	CO-CO	1965	8.23	Belyaev et al. (1967).
Ar-O ₂	1360	8.34	2.01 -2.50	Ar-Ar	171	6.06	Kamnev and Leonas (1965 a).
	5000	9.9	2.15 -2.63	Direct measurement			Jordan et al. (1970).
Ar-SF ₆	24.5 × 10 ⁶	12.8	3.24 -4.04	He-SF ₆	1.86 × 10 ⁹	11.48	Amdur (1967).
				He-Ar	62.1	7.25	Amdur et al. (1954).
				He-He	4.71	5.94	Amdur and Harkness (1954).
H ₂ -Xe	468.5	7.08 ₅	2.31 -2.76 ₅	He-H ₂	12.11	6.07	Amdur and Smith (1968).
				Xe-Xe	7.05 × 10 ³	7.97	Amdur and Mason (1956 a).
				He-He	4.71	5.94	Amdur and Harkness (1954).
H ₂ -CH ₄	51.7	4.04 ₅	1.84 -2.67	He-H ₂	5	3.8	Belyaev and Leonas (1967 b).
				Xe-Xe	463	6.35	Kamnev and Leonas (1966 a).
				He-He	4.33	5.86	Belyaev and Leonas (1967 b).
H ₂ -O ₂	1548	9.56	2.09 -2.54	He-CH ₄	602	9.43	Amdur et al. (1961 b).
				He-H ₂	12.11	6.07	Amdur and Smith (1968).
				He-He	4.71	5.94	Amdur and Harkness (1954).
H ₂ -O ₂	265	7.16	1.81 -1.99	Ar-O ₂	1360	8.34	Jordan et al. (1970).
				He-H ₂	12.11	6.07	Amdur and Smith (1968).
				He-Ar	62.1	7.25	Amdur et al. (1954).
H ₂ -SF ₆	58.2	6.1	1.84 -2.55	H ₂ -H ₂	14.1	5.87	Belyaev and Leonas (1967 b).
				O ₂ -O ₂	240	6.3	Belyaev and Leonas (1967 a).
				He-SF ₆	1.86 × 10 ⁹	11.48	Amdur (1967).
CH ₄ -He	4.78 × 10 ⁵	11.61	3.04 -3.53	He-H ₂	12.11	6.07	Amdur and Smith (1968).
				He-He	4.71	5.94	Amdur and Harkness (1954).
				He-He	4.71	5.94	Amdur and Harkness (1954).
CH ₄ -N ₂	602	9.43	1.92 -2.37	Direct measurement			Amdur et al. (1961 b).
				Ar-N ₂	755	7.78	Amdur et al. (1957).
CH ₄ -N ₂	832	7.30	2.41 -2.80	Ar-CH ₄	936	7.85	Mason and Amdur (1964).
				Ar-Ar	849	8.33	Amdur and Mason (1954).
				Ar-CH ₄	936	7.85	Mason and Amdur (1964).
CH ₄ -O ₂	1500	7.86	2.14 -2.47	Ar-O ₂	1360	8.34	Jordan et al. (1970).
				Ar-Ar	849	8.33	Amdur and Mason (1954).
				He-SF ₆	1.86 × 10 ⁹	11.48	Amdur (1967).
CH ₄ -SF ₆	28.0 × 10 ⁵	12.08	3.54 -3.75	Ar-CH ₄	936	7.85	Mason and Amdur (1964).
				He-Ar	62.1	7.21	Amdur et al. (1954).
				He-He	4.71	5.94	Amdur and Harkness (1954).
Ne-N ₂	605	9.08 ₅	2.03 ₅ -2.56	Ne-Ne	312	9.99	Amdur and Mason (1955 a).
				Ne-Ne	78	7.65	Kamnev and Leonas (1965 a).
				N ₂ -N ₂	550	7.4	Belyaev and Leonas (1967 a).
N ₂ -Kr	207	7.52 ₅	2.02 -2.61 ₅	He-N ₂	74.3	7.06	Amdur et al. (1957).
				Kr-Kr	159	5.42	Amdur and Mason (1955 b).
				He-He	4.71	5.94	Amdur and Harkness (1954).

TABLE 23. Molecular-beam potentials, $\varphi(r) = K/r^s$, for Group III^{a, b}—Continued

System	Potential			Source			Reference
	K, eV(Å) ^s	s	Range, Å	System	K, eV(Å) ^s	s	
N ₂ -Xe	872	7.55	2.37–3.03 ₅	N ₂ -N ₂	550	7.4	Belyaev and Leonas (1967 a).
	2874	8.07 ₅	2.66–3.29 ₅	Kr-Kr	1382	7.7	Kamnev and Leonas (1966 a).
				He-N ₂	74.3	7.06	Amdur et al. (1957).
				He-He	4.71	5.94	Amdur and Harkness (1954).
N ₂ -O ₂	505	6.87 ₅	2.41–3.07	Xe-Xe	7050	7.97	Amdur and Mason (1956 a).
	1630	8.15	2.16–2.52	N ₂ -N ₂	550	7.4	Belyaev and Leonas (1967 a).
				Xe-Xe	463	6.35	Kamnev and Leonas (1966 a).
				Ar-O ₂	1360	8.34	Jordan et al. (1970).
N ₂ -SF ₆	350	6.8	2.94–3.05	He-N ₂	74.3	7.06	Amdur et al. (1957).
	29.3 × 10 ⁵	12.60	3.39–4.06	He-Ar	62.1	7.25	Amdur et al. (1954).
				Direct measurement			Belyaev and Leonas (1967 a).
				He-SF ₆	1.86 × 10 ⁵	11.48	Amdur (1967).
CO-Kr	238	5.53 ₅	2.21–2.90 ₅	He-N ₂	74.3	7.06	Amdur et al. (1957).
	1648	7.96 ₅	2.35 ₅ –2.97	He-He	4.71	5.94	Amdur and Harkness (1954).
				Ar-CO	551	6.99	Jordan et al. (1970).
				Ar-Ar	849	8.33	Amdur and Mason (1954).
CO-O ₂	883	7.00	1.92–2.49	Kr-Kr	159	5.42	Amdur and Mason (1955 b).
	687	7.26 ₅	2.32 ₅ –3.03 ₅	CO-CO	1965	8.23	Belyaev et al. (1967).
				Kr-Kr	1382	7.7	Kamnev and Leonas (1966).
				CO-Ar	551	6.99	Jordan et al. (1970).
CO-SF ₆	16.5 × 10 ⁵	11.22	3.32–3.77	O ₂ -Ar	1360	8.34	Jordan et al. (1970).
	40.7 × 10 ⁵	12.53	3.24–3.59	Ar-Ar	849	8.33	Amdur and Mason (1954).
				CO-CO	1965	8.23	Belyaev et al. (1967).
				O ₂ -O ₂	240	6.3	Belyaev and Leonas (1967 a).
O ₂ -SF ₆	1.86 × 10 ⁵	11.48	2.87–3.36	He-SF ₆	1.86 × 10 ⁵	11.48	Amdur (1967).
	40.7 × 10 ⁵	12.53	3.24–3.59	Ar-CO	551	6.99	Jordan et al. (1970).
				He-Ar	62.1	7.25	Amdur et al. (1954).
				He-SF ₆	1.86 × 10 ⁵	11.48	Amdur (1967).
He-SF ₆	1.86 × 10 ⁵	11.48	2.87–3.36	Ar-O ₂	1360	8.30	Jordan et al. (1970).
	40.7 × 10 ⁵	12.53	3.24–3.59	He-Ar	62.1	7.25	Amdur et al. (1954).
				Direct measurement			Amdur (1967).
				Direct measurement			

^a Potentials were not determined for air-(Ar, CH₄, CO, SF₆) and CO₂-(Ar, CO, O₂, air, N₂O, SF₆) because molecular-beam measurements were unavailable.

^b Complete reference information is given in Bibliography II.

Ar-CO. The consistency of the closed-tube measurement by Ivakin and Suetin (1964 a) and \mathcal{D}_{12} by molecular-beam measurements with the results for Ar-N₂ (Group II) were the bases for placing Ar-CO into Group III.

Ar-O₂. This gas pair has only one set of direct measurements of \mathcal{D}_{12} available, obtained by the two-bulb method (Paul and Srivastava, 1961 a).

Ar-air. There are no direct measurements available, and the results were calculated by Blanc's law.

Ar-CO₂. The more reliable measurements for Ar-CO₂ are by Ivakin and Suetin (1964 b) and by Pakurar and Ferron (1964, 1966). The results by Pakurar and Ferron appear to have an unusual amount of scatter, which is due to difficult point-source measurements at temperatures above 1000 K.

Ar-SF₆. This gas pair has only one set of direct measurements of \mathcal{D}_{12} , obtained by the closed-tube method (Ivakin and Suetin, 1964 b).

H₂-Xe. The two-bulb measurements by Paul and Srivastava (1961 c) and the \mathcal{D}_{12} from mixture viscosity data calculated by Weissman and Mason (1962 b) are considered equally reliable.

H₂-CH₄. The closed-tube measurements by Boyd et al. (1951) and by Arnold and Toor (1967) agree within about 1 percent. These results are at room temperature; values of \mathcal{D}_{12} at temperatures up to 523 K were obtained from mixture viscosity data by Weissman and Mason (1962 b).

H₂-O₂. The reference equation essentially splits the difference between the high-temperature results by Walker and Westenberg (1960) and by Weissman and Mason (1962 b). For this gas pair the usually reliable point-source results by Walker and

Westenberg are considered possibly somewhat high. This conclusion is based on a comparison with the results of H₂-N₂ (Group I), which are expected to be similar. Spontaneous ignition occurred at about 920 K in the point-source measurements.

H₂-SF₆. The most reliable measurement for this gas pair is by Boyd et al. (1951); other reliable determinations by the closed-tube method are by Strehlow (1953) and by Ivakin and Suetin (1964 a, b).

CH₄-He. The reliable direct measurements of \mathcal{D}_{12} are primarily from recent open-tube studies by Frost (1967) and by Rhodes and Amick (1967).

CH₄-N₂. The two-bulb measurements by Mueller and Cahill (1964) were considered sufficient to place this gas pair into Group III.

CH₄-O₂. The only direct measurements available are those of Walker and Westenberg (1960) by the point-source technique. Spontaneous ignition occurred at about 1020 K. The results are not inconsistent with those of the similar system CH₄-N₂.

CH₄-air. There are no direct measurements available, and the results were calculated by Blanc's law.

CH₄-SF₆. This gas pair has only one set of direct measurements of \mathcal{D}_{12} , obtained by the closed-tube method (Manner, 1967).

N₂-Ne. This gas pair has reliable values of \mathcal{D}_{12} only from mixture viscosity measurements by DiPippo et al. (1967).

N₂-Kr. This gas pair has only one set of direct measurements of \mathcal{D}_{12} , obtained by the two-bulb method (Durbin and Kobayashi, 1962).

N₂-Xe. This gas pair has only one set of direct measurements of \mathcal{D}_{12} , obtained by the two-bulb method (Paul and Srivastava, 1961 b).

N₂-O₂. The most reliable results are the closed-tube measurements by Lonius (1909) and the values calculated from mixture viscosity by Weissman and Mason (1962 b). The results reported by Giddings and Seager (1962) are omitted from the deviation plot, figure 63, because of difficulties with the mixture composition analysis.

N₂-SF₆. The most reliable direct measurements are by Ivakin and Suetin (1964 b), obtained by the closed-tube method.

CO-Kr. This gas pair has only one set of direct measurements of \mathcal{D}_{12} , obtained by the two-bulb method (Singh et al., 1967). Since CO and N₂ are isosteric molecules, the reference equation for N₂-Kr was used for CO-Kr, and the data agree, see figure 65.

CO-O₂. Since CO and N₂ are isosteric molecules, the reference equation for N₂-O₂ was used for CO-O₂. The most reliable results are considered to be from mixture viscosity (Weissman and Mason, 1962 b). However, this judgment implies that the usually more reliable measurements by Loschmidt (1870 b) and by Walker and Westenberg (1960) are somewhat high.

CO-air. There are no direct measurements available, and the results were calculated by Blanc's law.

CO-CO₂. Since CO and N₂ are isosteric molecules, the reference equation for N₂-CO₂ (Group II) can be used for CO-CO₂; a slightly more precise correlation, however, is given for CO-CO₂ based only on its direct measurements. The direct measurements for CO-CO₂ are in the temperature range of 282 to 473 K. The use of the N₂-CO₂ reference equation will extend the higher temperature limit to 1800 K, which is a significant advantage.

CO-SF₆. This gas pair has direct measurements of \mathcal{D}_{12} which were obtained by the closed-tube method (Ivakin and Suetin, 1964 a, b).

O₂-CO₂. The reference equation is based primarily on results of point-source measurements (Walker and Westenberg, 1960); at room temperature the results by closed-tube studies give slightly higher values of \mathcal{D}_{12} (Loschmidt, 1870 b; Wretschko, 1870).

O₂-SF₆. This gas pair has reliable closed-tube measurements by Ivakin et al. (1968), which, however, probably have somewhat too great a temperature dependence for \mathcal{D}_{12} over 297 to 408 K.

CO₂-air. Even though there are many direct measurements available, the reference equation for CO₂-air was calculated from Blanc's law. Of the direct measurements, the most reliable are considered to be closed-tube measurements by Loschmidt (1870 a, b), by Coward and Georgeson (1937), and by Holsen and Strunk (1964). The open-tube measurements by Klivanova et al. (1942), which cover the temperature range of 290 to 1533 K, are not considered as reliable as results by Blanc's law with data from the point-source method measurements by Walker (1958) and by Pakurar and Ferron (1964, 1966) for N₂-CO₂, and by Walker and Westenberg (1960) for O₂-CO₂.

CO₂-N₂O. This gas pair has several closed-tube measurements which agree within about 2 percent at room temperature (Loschmidt, 1870 b; Boardman and Wild, 1937; Wall and Kidder, 1946; Amdur et al., 1952); values of \mathcal{D}_{12} from mixture viscosity were used to extend the temperature range to 550 K. The placement of CO₂-N₂O into Group III, and not Group II, was decided upon because of the limited tem-

perature range of the direct measurements and the uncertainties in \mathcal{D}_{12} obtained from mixture viscosity for cylindrical molecules.

CO₂-SF₆. This gas pair has reliable closed-tube measurements by Ivakin and Suetin (1964 b).

SF₆-He. This gas pair has a few direct measurements, of which those by Ivakin and Suetin (1964 b) and by Fedorov et al. (1966) are considered to be the most reliable.

SF₆-air. There are no direct measurements available, and the results were calculated from Blanc's law.

d. Miscellaneous (Figs. 76 to 81)

Weights and Potentials. The values of \mathcal{D}_{12} and their weights used in the least-squares calculations are presented in table 24. Except for mixtures with dissociated gases the values of \mathcal{D}_{12} are at equimolar composition. The data for mixtures with dissociated gases were not corrected to equimolar composition because the experimental uncertainties are greater than the composition dependence of \mathcal{D}_{12} . The potential functions obtained from molecular-beam measurements, which are summarized in table 25, are available only for the mixtures with dissociated gases and not the other systems of the miscellaneous group. This information may be helpful for the prediction of \mathcal{D}_{12} at high temperatures, but the potentials were not used to calculate points for deviation plots as was done for the other gas pairs in Groups I, II, and III. There are no deviation plots for mixtures with dissociated gases because of the large uncertainties in the data available.

Special Comments. For the fourteen gas pairs of the miscellaneous group the special comments are as follows.

H₂O-N₂. This gas pair has direct measurements obtained only by the evaporation-tube method. The most reliable results are considered to be by O'Connell et al. (1969), in which the H₂O diffused downwards through N₂. In the other studies the H₂O was located below the N₂, or the lighter component below the heavier, which would have possible adverse effects due to convection. Because values of \mathcal{D}_{12} are available only from evaporation-tube studies, it may be noted that the temperature range is limited, 282 to 373 K.

H₂O-O₂. The correlation consists of two power functions ($\mathcal{D}_{12}=AT^s$), each applicable over a specific temperature interval, and which pass through a common datum. The generation of this type of correlation is explained as follows. At high temperatures, 390 to 1070 K, the point-source measurements are considered to be reliable (Walker and Westenberg, 1960); however at low temperatures, 308 to 352 K, the H₂O-O₂ evaporation-tube studies are considered too uncertain. But, at low temperatures, data by O'Connell et al. (1969) for H₂O-N₂ were systematically increased by 1 percent to obtain values of \mathcal{D}_{12} for H₂O-O₂. This slight adjustment was based on calculations for transport properties of polar-gas mixtures [3]. But all results for H₂O-O₂ could not be well correlated by a single equation, either in the form of a Sutherland equation, eq (4.3-2), or the more complex correlation function of eq (4.3-1). The simplest reliable curve-fit of the data is two power functions, one each for the low- and high-temperature ranges. The equation at high temperatures was obtained by least-squares calculations of the point-source measurements. This equation was

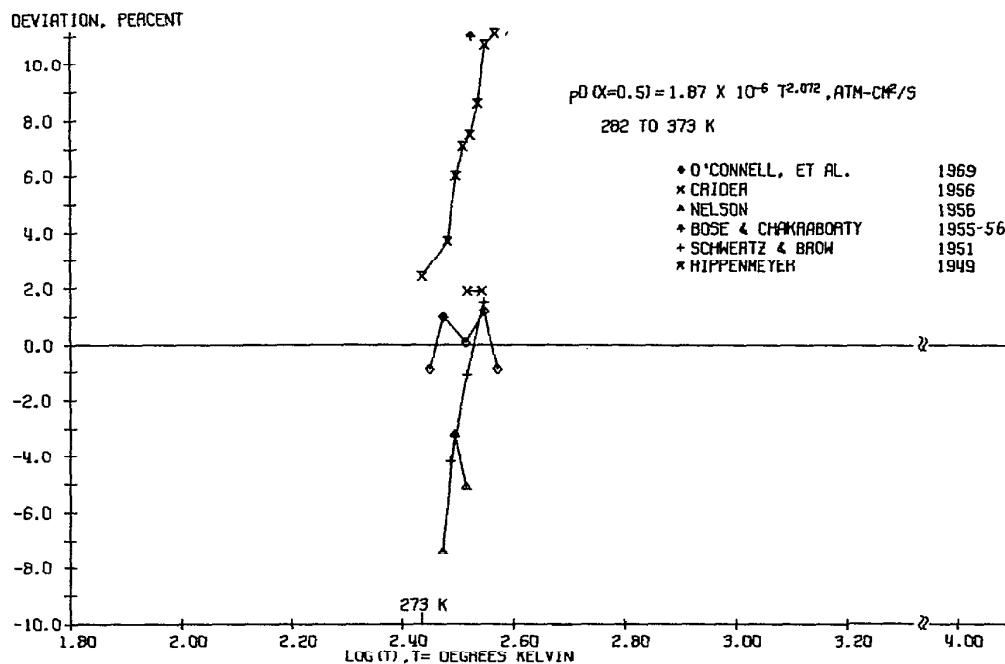


FIGURE 76. Deviations of diffusion coefficients from reference equation.

Water-Nitrogen

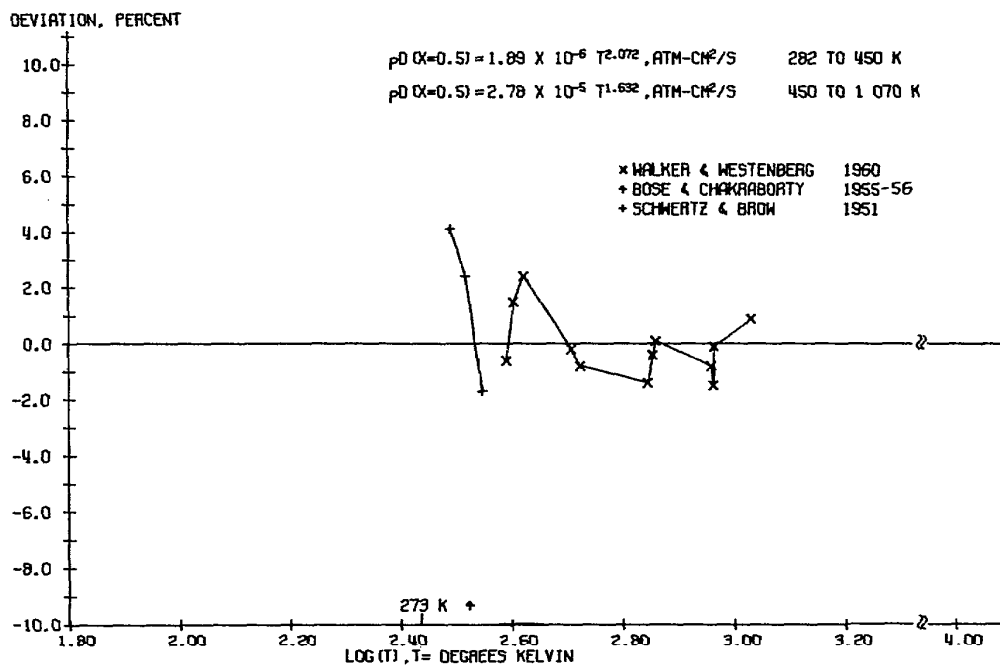


FIGURE 77. Deviations of diffusion coefficients from reference equation.

Water-Oxygen

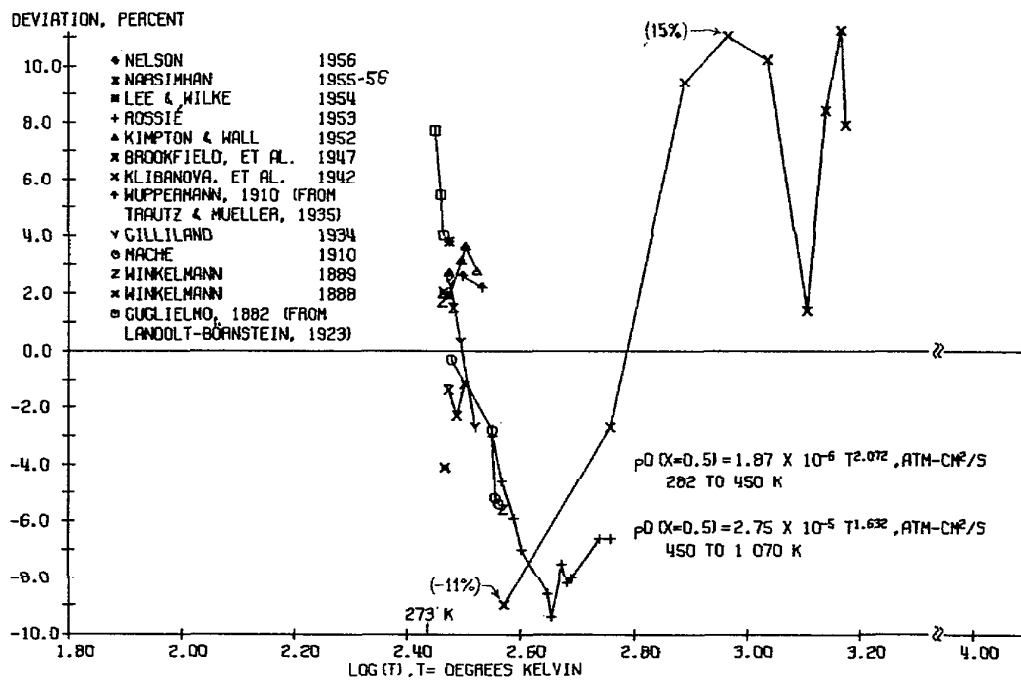


FIGURE 78. Deviations of diffusion coefficients from reference equation.

Water-Air

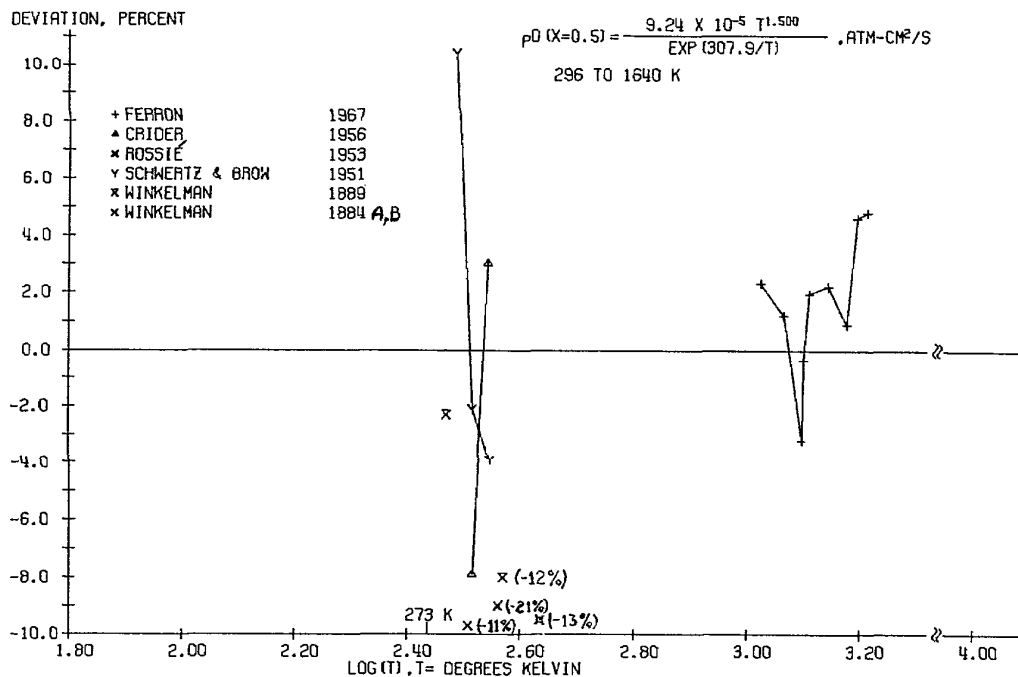


FIGURE 79. Deviations of diffusion coefficients from reference equation.

Water-Carbon dioxide

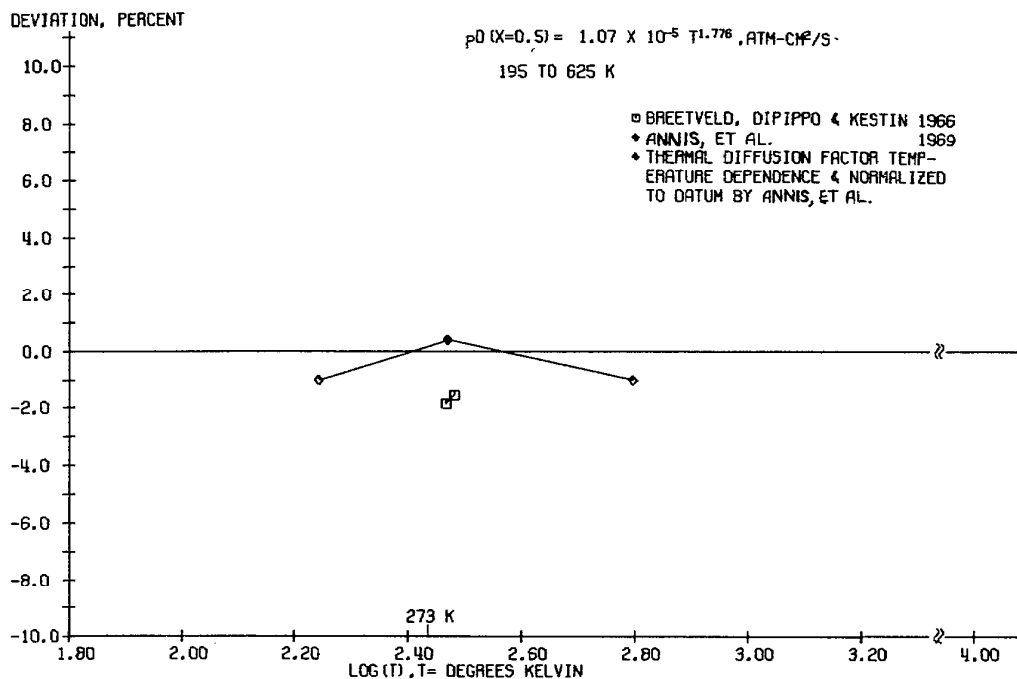


FIGURE 80. Deviations of diffusion coefficients from reference equation.

Neon-Carbon dioxide

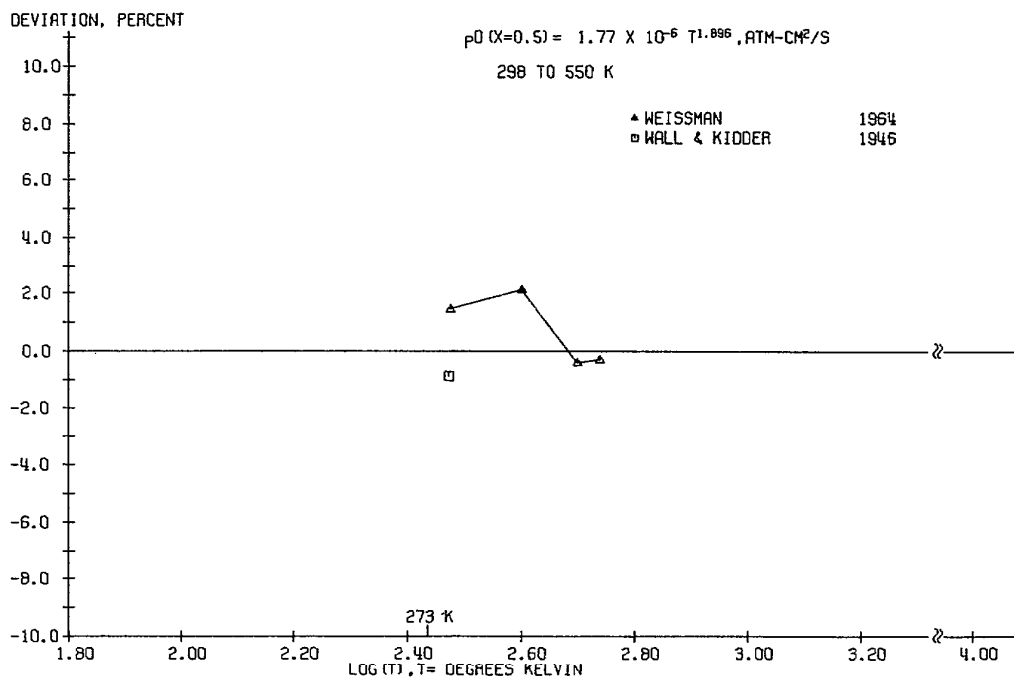


FIGURE 81. Deviations of diffusion coefficients from reference equation.

Propane-Carbon dioxide

forced to pass through a point at the intermediate temperature of 450 K, obtained from an extrapolation of the low-temperature equation.

H₂O-air. The reference equation for this air-system is the only one in this report which was not determined by Blanc's law. The reason is that the available data for H₂O-N₂ and H₂O-O₂ have too much scatter, and a verification of a correlation by Blanc's law from direct measurements for H₂O-air would not be significant. The reference equation for H₂O-air is based on the synthesis of results by O'Connell et al. (1969) for H₂O-N₂ and by Walker and Westenberg (1960) for H₂O-O₂. Since air is approximately 80 percent N₂, the reference equation for H₂O-N₂ extrapolated to 450 K, was assumed directly applicable at low temperatures. For temperatures between 450 and 1070 K, the reference equation for H₂O-O₂ was systematically reduced 1 percent. The more reliable direct measurements are shown in figure 78. The large deviations are due to expected uncertainties in results obtained by evaporation-tube studies. There are many other experimental determinations for H₂O-air, which are listed in table 16.

H₂O-CO₂. This gas pair has reliable direct measurements by Ferron (1967), obtained by the point-source method over the temperature range of 1058 to 1640 K, and evaporation-tube studies at about 310 to 350 K, obtained by Schwertz and Brow (1951) and by Crider (1956). In the least-squares calculations the value of *s* of eq (4.3-2) turned out to be 1.473 which was adjusted to 1.500 to agree with the theoretical lower limit for the rigid-sphere model.

CO₂-Ne. This gas pair has limited data, consisting of a two-bulb measurement at about room temperature plus information on the temperature dependence of the thermal diffusion factor which was used to calculate \mathcal{D}_{12} between 195 to 625 K.

CO₂-C₃H₈. The closed-tube measurements by Wall and Kidder (1946) and the values of \mathcal{D}_{12} from mixture viscosity by Weissman (1964) are considered sufficient to include this system.

Mixtures with dissociated gases were all correlated by power functions of the form $\mathcal{D}_{12} = AT^s$. The reference equations were calculated from two points, one at about room temperature, and the other at temperatures greater than 1000 K. Deviation plots are not given for mixtures with dissociated gases. The special comments emphasize the discrepancies among the various values of \mathcal{D}_{12} obtained by different experiments.

H-He. This gas pair has only one direct measurement by Khouw et al. (1969), obtained at 275 K. The values of \mathcal{D}_{12} obtained from molecular-beam measurements by Amdur and Mason (1956 b) and by Belyaev and Leonas (1967 b, c) were essentially averaged; the difference in \mathcal{D}_{12} between these results is approximately 25 percent at all temperatures.

H-Ar. The direct measurements by Wise (1959) and by Khouw et al. (1969), near room temperature, differ by about 10 percent. At elevated temperatures, values of \mathcal{D}_{12} are available only from one potential (Mason and Vanderslice, 1958), and these results when compared with the reference equation are high by about 25 percent at 2000 K and low by about 20 percent at 10 000 K.

TABLE 24. Diffusion coefficients and weights for curve-fitting, miscellaneous group^a

System	T, K	log ₁₀ [$\mathcal{D}_{12}(x=1/2)$]	Note	System	T, K	log ₁₀ [$\mathcal{D}_{12}(x=1/2)$]	Note
H ₂ O-N ₂	281.9	-0.6554	b	H ₂ O-CO ₂	1200	0.4639	h
	298.2	-.5965	b		1300	.5198	h
	327.5	-.5158	b		1400	.5775	h
	327.5	-.5200	b		1500	.6325	h
	327.4	-.5131	b		1600	.6884	h
H ₂ O-O ₂	353.2	-.4436	b	1700	.7482	h	
	373.4	-.4029	b	CO ₂ -Ne	175	-.9914	i
	450.0	-.2261	c		625	-.0097	i
	511	-.1367	d	CO ₂ -C ₃ H ₈	298.1 ₅	-1.0665	j
	529	-.1146	d		298.1 ₅	-1.0620	j
	698	+0.0792	d		300.0	-1.0492	k
	715	.1007	d		400.0	-.08097	k
	722	.1096	d		500.0	-.6364	k
	908	.2683	d	550.0	-.5575	k	
	917	.2721	d	H-He	275	.3766	l
	921	.2813	d		4620	2.500	*
1069	.3879	d	H-Ar	282	0.075	*	
1069	.3939	d		4620	2.015	*	
1070	.3928	d	H-H ₂	274	0.2667	m	
H ₂ O-air	-	-		10 000	2.966	*	
H ₂ O-CO ₂	307.4 ₅	-.6947	f	N-N ₂	280	-0.5376	n
	328.5 ₃	-.6757	f				
	352.3 ₃	-.6108	f	O-N ₂	2850	1.250	*, n
	328.6 ₅	-.7033	g				
	349.1 ₅	-.5901	g	O-He	316	0.043	*
	1000	.3617	h		10 000	2.6665	o
	1100	.4099	h	O-Ar	316	-.0522	*
					3760	1.457	*

^aSelected value, see explanation in first part of section 5.4.

^bAll listed values of \mathcal{D}_{12} are weighted one; except in H₂O-CO₂ for which the data between 1000 and 1700 K inclusive, are weighted two. For these systems, in almost all instances, the composition dependence of \mathcal{D}_{12} is insignificant; maximum correction is 0.45 percent for H₂O-O₂ datum at 1070 K.

^cO'Connell et al. (1969).

^dCommon point for low- and high-temperature correlations, see text (Section 5.4, part d).

^eWalker and Westenberg (1960, 1966).

^fSee section 5.4, part d.

^gSchwertz and Brow (1951).

^hCrider (1956).

ⁱFerron (1967); weight of datum doubled for least-squares calculations.

^jCalculated from data on temperature dependence of thermal diffusion factor (Weissman et al., 1961) and method of Annis et al. (1968). The reference equation lower temperature limit is reported as 195 K which is in agreement with the correct temperature limit of the thermal diffusion factor data; the equation was verified to be correct, even though a value of *T* of 175 K was used in the curve-fitting calculations.

^kWall and Kidder (1946).

^lWeissman (1964).

^mKhouw et al. (1969).

ⁿBrowning and Fox (1964).

^oMorgan and Schiff (1964), average of results for N-N₂, O-N₂, and O-O₂.

^pCalculated from potential of Leonas et al., see table 25.

TABLE 25. *Molecular-beam potentials, $\phi(r) = K/r^s$, for miscellaneous group^{a,b}*

System	Potential			Source			Reference
	K, eV(Å) ^s	s	Range, Å	system	K, eV(Å) ^s	s	
H-He	2.34	3.29	1.16-1.71	Direct measurement			Amdur and Mason (1956 b).
	1.2	2.7	0.79-1.35	Direct measurement			Belyaev and Leonas (1967 b).
H-Ar	31.6	4.49	2.1-3.0	As reported			Mason and Vanderslice (1958).
	6.26	1.99	1.32-1.88	He-H	1.2	2.7	Belyaev and Leonas (1967 b).
H-H ₂	6.02	3.42	1.33-1.88	He-Ar	22.6	5.15	Kamnev and Leonas (1965 a).
				He-He	4.33	5.86	Belyaev and Leonas (1967 b).
N-N ₂	76.6	6.31	1.76-2.54	He-H	2.34	3.29	Amdur and Mason (1956 b).
				He-H ₂	12.11	6.07	Amdur and Smith (1968).
O-He	38.0	7.99	1.20-1.60	He-He	4.71	5.94	Amdur and Harkness (1954).
				Direct measurement			Belyaev and Leonas (1967 b).
O-Ar	239	8.09	1.78-2.40	Direct measurement			Belyaev and Leonas (1966 b).
				Ar-O	239	8.09	Belyaev et al. (1967).
O-N ₂	22.5	5.0	2.00-2.40	He-Hc	4.33	5.86	Belyaev and Leonas (1967 b).
				Ar-Ar	171	6.06	Kamnev and Leonas (1965 a).
O-O ₂	13.25	4.4	2.05-2.46	Direct measurement			Belyaev et al. (1967).
				Direct measurement			Belyaev and Leonas (1966 b).

^aPotentials were not determined for H₂O-(N₂, O₂, air, CO₂) and CO₂-(N₂O, C₂H₆) because molecular-beam measurements were unavailable.
^bComplete reference information is given in Bibliography II.

H-H₂. For this gas pair the most reliable values of \mathcal{D}_{12} are considered to be from mixture viscosity (Browning and Fox, 1964). The other determinations of \mathcal{D}_{12} are considered to be less reliable (Wise, 1961; Weissman and Mason, 1962 a; Khouw et al., 1969; Sancier and Wise, 1969). The relative measurements by Wise (1961) are at temperatures from 293 to 719 K. At room temperature, the discrepancies are within ± 10 percent. The recommended values of \mathcal{D}_{12} above 1000 K are based on molecular-beam measurements. These results were derived from measurements by Amdur et al. and use of the combination rules, which were considered to be much more reliable than the direct beam measurement by Belyaev and Leonas (1967 b, c). The latter yield \mathcal{D}_{12} which are too high in comparison with both the low-temperature data, and results of other beam measurements.

N-N₂, O-N₂, O-O₂. For these gas pairs, with similar diffusion characteristics, the results by Morgan and Schiff (1964) are considered to be the most re-

liable. For O-O₂, at about room temperature, the measurements by Krongelb and Strandberg (1959) and by Walker (1961) are within 10 percent (below) those by Morgan and Schiff; the measurements by Yolles and Wise (1968) and by Yolles et al. (1970) are about 20 percent below those of Morgan and Schiff. The results by Walker may be low due to the neglect of chemical reaction effects. At temperatures between 1000 and 10 000 K the differences between \mathcal{D}_{12} from molecular-beam measurements for N-N₂, O-N₂, O-O₂ (Belyaev and Leonas, 1966 c) were so small that these results were grouped together.

O-He, O-Ar. There are two direct measurements for each of these gas pairs. In comparison with the results by Morgan and Schiff (1964), which are considered the more reliable, the results by Yolles and Wise (1968) are low by about 35 percent for O-He, and high by about 30 percent for O-Ar. The values of \mathcal{D}_{12} at elevated temperatures were based on a single laboratory source for the potentials (Leonas et al.).

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted*

The order of listing in Table 16 is as follows: (i) mixtures of noble gases with noble gases arranged according to the atomic weight of the lighter component, (ii) mixtures of noble gases with other gases arranged according to the atomic weight of the noble gas component, (iii) dissociated gases, and (iv) other mixtures arranged according to the molecular weight of the lighter component.

a. Noble Gases		
System	Reference	T, K
³ He- ³ He	Luszczynski et al. (1962)	1.7 to 4.2
	Luszczynski et al. (1967)	1.13 to 4.22
³ He- ⁴ He	Bendt (1958)	1.74 to 296
	Weissman and Mason (1962 b)	2.64 to 4.25
	DuBro (1969)	77 to 344
	DuBro and Weissman (1970)	77 to 888
⁴ He- ⁴ He	Amdur and Mason (1958)	1000 to 15 000
	Srivastava and Barua (1959)	273 to 318
He-Ne	Holmes and Tempest (1960)	298
	Weissman and Mason (1962 b)	20 to 523
	Weissman (1965)	291 and 302
	DiPippo et al. (1967)	293 and 303

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

a. Noble Gases—Continued		
System	Reference	T, K
He-Ar	Malinauskas (1968)	273 to 394
	van Heijningen et al. (1968)	65 to 295
	DuBro (1969)	77 to 364
	Malinauskas and Silverman (1969)	273 to 394
	DuBro and Weissman (1970)	77 to 365
	Hogervorst (1971)	300 to 1400
	Schmidt (1904)	286 to 292
	Lonius (1909)	286 to 295
	Strehlow (1953)	288 to 418
	Schäfer and Moesta (1954)	200 to 400
	Amdur and Mason (1958)	1000 to 15 000
	Walker (1958)	298
	Saxena and Mason (1959)	251 to 418
	Srivastava (1959)	273 to 318
	Walker and Westenberg (1959)	298 to 1063
	Holmes and Tempest (1960)	298
	Evans et al. (1961)	293
	Mason (1961)	303
	Suetin and Ivakin (1961)	287
	Evans et al. (1962)	298 and 373
	Giddings and Seager (1962)	296
	Weissman and Mason (1962 b)	72 to 473
	Evans et al. (1963)	298
	Golubev and Bondarenko (1963)	298 and 363
	Seager et al. (1963)	298 to 498
	Holsen and Strunk (1964)	276 to 346
	Ivakin and Suetin (1964 b)	287 to 465
	Suetin (1964)	287
	Ljunggren (1965)	293
	Malinauskas (1965)	273 to 394
	Weissman (1965)	291 to 311
	Carey et al. (1966)	300
	Fedorov et al. (1966)	291
	Kosov and Karpushin (1966)	169 to 296
	Kosov and Karpushin (1966 a)	293
	Malinauskas (1966)	273 to 394
	Mason and Smith (1966)	334
	Coates and Mian (1967)	298 to 522
	DiPippo et al. (1967)	293 and 303
	Mian (1967)	298 to 522
	Carey et al. (1968)	300, 1255 to 4990
	Mathur and Saxena (1968)	270 to 350
	van Heijningen et al. (1968)	90 to 400
	Annis et al. (1969)	295
	DuBro (1969)	305 and 335
	Hawtin et al. (1969)	293 to 873
	Schneider and Schäfer (1969)	273 to 1300
Wasik and McCulloh (1969)	77 to 357	
DuBro and Weissman (1970)	305 to 335	
Hu and Kobayashi (1970)	248 to 323	
Kalelkar and Kestin (1970)	298 to 993	
Hogervorst (1971)	300 to 1400	
He-Kr	Srivastava and Barua (1959)	273 to 318
	Holmes and Tempest (1960)	298
	Durbin and Kobayashi (1962)	308
	Srivastava and Paul (1962)	305
	Weissman and Mason (1962 b)	291
	Watts (1964)	303
	Weissman (1965)	291 and 302
	Fedorov et al. (1966)	290
	Kestin et al. (1966)	293 and 303
	Malinauskas (1966)	273 to 394
	Mason and Smith (1966)	318
	Annis et al. (1968)	77 to 760
	van Heijningen et al. (1968)	112 to 400
	Wasik and McCulloh (1969)	298 to 366
	Kalelkar and Kestin (1970)	298 to 993
	Hogervorst (1971)	300 to 1100
	He-Xe	Srivastava (1959)
Weissman and Mason (1962 b)		291 to 550
Malinauskas (1965)		273 to 394
Watts (1965)		303
Weissman (1965)		291 to 311
He-Rn	van Heijningen et al. (1968)	169 to 400
	Hogervorst (1971)	300 to 1000
Ne-Ne	Hirst and Harrison (1939)	283–286
	Groth and Sussner (1944)	293
Ne-Ar	Winn (1950)	78 to 353
	Amdur and Mason (1958)	1000 to 15 000
	Schäfer and Schuhmann (1957)	90 to 473

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

a. Noble Gases—Continued				
System	Reference	T , K		
Ne-Kr	Srivastava and Srivastava (1959)	273 to 318		
	Weissman and Mason (1962 b)	72 to 523		
	Weissman (1965)	291 to 311		
	Freudenthal (1966)	300 to 600		
	DiPippo et al. (1967)	293 and 303		
	Hogervorst and Freudenthal (1967)	300 to 650		
	Malinauskas (1968)	273 to 394		
	van Heijningen et al. (1968)	90 to 400		
	Malinauskas and Silverman (1969)	273 to 394		
	Kestin et al. (1970)	298 to 973		
	Hogervorst (1971)	300 to 1400		
	Srivastava and Srivastava (1959)	273 to 318		
	Paul (1962)	302		
	Weissman and Mason (1962 b)	291		
	Watts (1964)	303		
Ne-Xe	Weissman (1965)	291 to 311		
	Malinauskas (1968)	273 to 394		
	Mathur and Saxena (1968)	270 to 350		
	van Heijningen et al. (1968)	112 to 400		
	Malinauskas and Silverman (1969)	273 to 394		
	Hogervorst (1971)	300 to 1400		
	Srivastava and Barua (1959)	273 to 318		
	Weissman and Mason (1962 b)	291		
	Watts (1965)	303		
	Weissman (1965)	291 to 302		
	Malinauskas (1968)	273 to 394		
	van Heijningen et al. (1968)	169 to 400		
	Weissman (1968 b)	328 to 873		
	DuBro (1969)	304		
	Malinauskas and Silverman (1969)	273 to 394		
Ne-Rn	Taylor et al. (1969)	328 to 873		
	Weissman (1969)	305 to 925		
	Weissman and DuBro (1970 a)	304 to 922		
	Hogervorst (1971)	300 to 1400		
	Hirst and Harrison (1939)	290 and 293		
	Ar-Ar	Hutchinson (1947)	295	
		Hutchinson (1949)	90 to 327	
		Winn (1950)	78 to 353	
		Amdur and Schatzki (1957)	273	
		Amdur and Mason (1958)	1000 to 15 000	
		De Paz et al. (1967)	76 to 294	
		Vugts et al. (1969)	235 to 418	
		Ar-Kr	Schäfer and Schuhmann (1957)	200 to 473
			Srivastava and Srivastava (1959)	273 to 318
			Durbin and Kobayashi (1962)	248 to 308
Paul (1962)			302	
Weissman and Mason (1962 b)			291	
Watts (1964)			303	
Weissman (1965)			291 to 311	
Fedorov et al. (1966)			291	
Malinauskas (1966)	273 to 394			
Ivakin et al. (1968)	297 to 407			
van Heijningen et al. (1968)	169 to 400			
Humphreys and Mason (1970)	77 to 600			
Kestin et al. (1970)	298 to 973			
Hogervorst (1971)	300 to 1400			
Ar-Xe	Amdur and Schatzki (1957)		195 to 378	
	Amdur and Schatzki (1958)	330		
	Srivastava (1959)	273 to 318		
	Weissman and Mason (1962 b)	291		
	Malinauskas (1965)	273 to 394		
	Watts (1965)	303		
	Weissman (1965)	291 to 311		
	van Heijningen et al. (1968)	169 to 400		
	Hogervorst (1971)	300 to 1400		
	Ar-Rn	Hirst and Harrison (1939)	282 and 286	
		Kr-Kr	Groth and Harteck (1941)	294 and 296
			Schäfer and Schuhmann (1957)	199 to 474
			Amdur and Mason (1958)	1000 to 15 000
			Miller and Carman (1961)	293
			Durbin and Kobayashi (1962)	308
Paul (1962)			302	
Srivastava and Paul (1962)			305	
Wendt et al. (1963)			232 to 470	
Miller and Carman (1964)			293	
Watts (1964, 1965)			303	
Kamnev and Leonas (1966)			2000 to 10 000	
Saran and Singh (1966)			303	
Annis et al. (1969)			295	
DuBro (1969)			305 and 367	
Weissman and DuBro (1970 b)	196 to 1036			

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

a. Noble Gases—Continued			
System	Reference	T, K	
Kr-Xe	Weissman and Mason (1962 b)	291	
	Watts (1964, 1965)	303	
	Weissman (1965)	291 and 302	
	Malinauskas (1966)	273 to 394	
Xe-Xe	van Heijningen et al. (1968)	169 to 400	
	Groth and Harteck (1941)	292	
	Visner (1951 a, b)	300	
	Amdur and Schatzki (1957)	195 to 378	
	Amdur and Mason (1958)	1000 to 15 000	
	Watts (1965)	303	
	Kamnev and Leonas (1966)	2000 to 10 000	
b. Noble Gases and Another Component			
System	Reference	T, K	
He-H ₂	Bunde (1955)	298	
	Rumpel (1955)	298 to 358	
	van Itterbeek and Nihoul (1957)	52 to 153	
	Suetin et al. (1960)	273	
	Suetin and Ivakin (1961)	292	
	Giddings and Seager (1962)	298	
	Weissman and Mason (1962 b)	90 to 523	
	Suetin (1964)	294	
	Amdur and Malinauskas (1965)	195 to 374	
	Giddings (1968)	298	
	Kestin and Yata (1968)	293 and 303	
	Amdur and Malinauskas (1965)	195 to 374	
	He-D ₂	Ivakin and Suetin (1964 a)	295
	He-T ₂	Amdur and Malinauskas (1965)	195 to 374
He-CH ₄	Carswell and Stryland (1963)	298	
He-NH ₃	Fuller and Giddings (1965)	373	
	Arai et al. (1967)	313	
	Frost (1967)	303 to 764	
	Rhodes and Amick (1967)	302 to 627	
	Hu and Kobayashi (1970)	248 to 323	
	Giddings and Seager (1962)	297	
	Srivastava (1962)	274 to 333	
	Ivakin and Suetin (1964 a)	297	
	He-H ₂ O	Schwartz and Brow (1951)	307 to 352
	Lee and Wilke (1954)	298	
He-Na	von Hartel et al. (1932)	655	
	Anderson and Ramsey (1963)	427	
	Ramsey and Anderson (1964)	428	
	Cozzini et al. (1967)	273 and 443	
He-C ₂ H ₂	Violino (1968)	427 to 443	
	Suetin and Ivakin (1961)	290	
He-C ₂ H ₄	Suetin (1964)	290	
	Frost (1967)	303 to 765	
He-N ₂	Rumpel (1955)	298 to 358	
	Westenberg and Walker (1957)	293	
	Walker (1958)	297 to 1124	
	Walker and Westenberg (1958 a, b)	298 to 1200	
	Paul and Srivastava (1961 b)	243 to 333	
	Suetin and Ivakin (1961)	289	
	Giddings and Seager (1962)	296 to 304	
	Seager et al. (1963)	298 to 498	
	Ivakin and Suetin (1964 b)	289 to 470	
	Suetin (1964)	289	
	Chang (1966)	244 to 311	
	Kestin et al. (1966)	293 and 303	
	Walker and Westenberg (1966)	297 to 1124	
	Coates and Mian (1967)	299 to 500	
	Frost (1967)	303	
	Henry et al. (1967)	299	
	Mian (1967)	299 to 500	
	Zhukhovitskiĭ et al. (1968)	293	
	Ellis and Holsen (1969)	297 to 882	
	Hawtin et al. (1969)	293 to 873	
	Wasik and McCulloh (1969)	77 to 370	
	Hu and Kobayashi (1970)	248 to 323	
	Nagata and Hasegawa (1970)	310 to 360	
He-CO	Ivakin and Suetin (1964 a)	296	
	Ivakin and Suetin (1964 b)	296 to 470	
He-C ₂ H ₆	Arai et al. (1967)	313	
	Frost (1967)	303 to 751	
He-O ₂	Kaufmann (1967)	373 to 523	
	Paul and Srivastava (1961 a)	244 to 334	
	Suetin and Ivakin (1961)	287	

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

b. Noble Gases and Another Component—Continued		
System	Reference	T, K
	Giddings and Seager (1962)	297
	Seager et al. (1963)	298 to 498
	Suetin (1964)	287
	Kestin and Yata (1968)	293 and 303
He-air	Wasik and McCulloh (1969)	298 to 365
	Suetin et al. (1960)	273
	Suetin and Ivakin (1961)	287
	Holsen and Strunk (1964)	276 to 346
	Ivakin and Suetin (1964 b)	287 to 469
	Suetin (1964)	287
	Fedorov et al. (1966)	292
He-CH ₃ OH	Evans et al. (1969)	295
He-K	Seager et al. (1963)	423 to 523
	Bernheim and Korte (1965)	358
	Khomchenkov et al. (1968)	723
	Violino (1968)	358
He-C ₃ H ₆	Ivanovskii et al. (1969)	570 to 930
	Weissman (1964)	293 to 523
	Frost (1967)	303 to 764
He-CO ₂	Lonsdale and Mason (1957)	260 to 358
	Saxena and Mason (1959)	250 to 404
	McCarty and Mason (1960)	303
	Suetin et al. (1960)	273
	Walker et al. (1960)	299
	Suetin and Ivakin (1961)	287
	Giddings and Seager (1962)	300
	Seager et al. (1963)	298 to 498
	Holsen and Strunk (1964)	276 to 346
	Ivakin and Suetin (1964 b)	287 to 465
	Suetin (1964)	287
	Kosov and Novosad (1966 a)	294
	DiPippo et al. (1967)	293 and 303
	Ferron and Dunham (1967)	782
	Oost et al. (1967)	295 and 343
	Kosov and Bogatyrev (1968)	290 to 430
	Annis et al. (1969)	295
	Hu and Kobayashi (1970)	248 to 323
	Nagata and Hasegawa (1970)	314 to 365
He-C ₃ H ₈	Frost (1967)	303 to 767
	Kaufmann (1967)	373 to 503
He-C ₂ H ₅ OH	Rhodes and Amick (1967)	303
	Lee and Wilke (1954)	298
	Seager et al. (1963)	423 to 523
He-difluoromethane	Hargrove and Sawyer (1967)	298
He-1-butene	Fuller et al. (1969)	431
He-2-butene	Frost (1967)	303 to 522
He-C ₄ H ₁₀	Weissman (1964)	293 to 523
	Frost (1967)	303 to 751
	Rhodes and Amick (1967)	303 to 477
	Hargrove and Sawyer (1967)	298 to 473
He-acetone	Hargrove and Sawyer (1967)	298 to 473
He-1-propanol	Seager et al. (1963)	423 to 523
He-2-propanol	Seager et al. (1963)	423 to 523
He-1,1-difluoroethane	Fuller et al. (1969)	430
He-n-pentane	Hargrove and Sawyer (1967)	298 to 473
He-ether	Hargrove and Sawyer (1967)	298 to 473
He-1-butanol	Seager et al. (1963)	423 to 523
He-benzene	Lee and Wilke (1954)	298
	Seager et al. (1963)	423 to 523
	Hargrove and Sawyer (1967)	298 to 473
He-1-chloropropane	Fuller et al. (1969)	428
He-dichloromethane	Fuller et al. (1969)	428
He-Rb	Bernheim (1962)	323
	Violino (1968)	323 and 340
He-3-pentanone	Barr and Sawyer (1964)	300
He-n-hexane	Hargrove and Sawyer (1967)	298 to 473
	Fuller and Giddings (1965)	417
He-1-pentanol	Seager et al. (1963)	423 to 523
He-1-chlorobutane	Fuller et al. (1969)	429
He-2-chlorobutane	Fuller et al. (1969)	429
He-fluorobenzene	Fuller et al. (1969)	430
He-1,2-dichloroethane	Fuller et al. (1969)	427
He-n-heptane	Clarke and Ubbelohde (1957)	303
He-2,4-dimethylpentane	Clarke and Ubbelohde (1957)	303
He-1-hexanol	Seager et al. (1963)	423 to 523
He-1-fluorohexane	Fuller et al. (1969)	432
He-1-chloropentane	Fuller et al. (1969)	428
He-bromoethane	Fuller et al. (1969)	428
He-4-fluorotoluene	Fuller et al. (1969)	432
He-chlorobenzene	Fuller et al. (1969)	431

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

b. Noble Gases and Another Component—Continued		
System	Reference	T, K
He- <i>n</i> -octane	Clarke and Ubbelohde (1957)	303
He-2,2,4-trimethylpentane	Clarke and Ubbelohde (1957)	303
He-trichloromethane	Fuller et al. (1969)	429
He-1-bromopropane	Fuller et al. (1969)	428
He-2-bromopropane	Fuller et al. (1969)	428
He-nitrobenzene	Lee and Wilke (1954)	298
He-Cs	Legowski (1964)	299
	Khomchonkov et al. (1968)	723
	Violino (1968)	299
He-1-bromobutane	Fuller et al. (1969)	427
He-2-bromobutane	Fuller et al. (1969)	427
He-iodomethane	Fuller et al. (1969)	431
He-SF ₆	Suetin and Ivakin (1961)	291
	Suetin (1964)	291
	Ivakin and Suetin (1964 b)	291 to 464
	Evans and Kenney (1965)	293
	Fedorov et al. (1966)	291
He-iodoethane	Fuller et al. (1969)	428
He-bromobenzene	Fuller et al. (1969)	427
He-2-bromo-1-chloropropane	Fuller et al. (1969)	427
He-1-bromohexane	Fuller et al. (1969)	428
He-2-bromohexane	Fuller et al. (1969)	428
He-3-bromohexane	Fuller et al. (1969)	429
He-1-iodopropane	Fuller et al. (1969)	430
He-2-iodopropane	Fuller et al. (1969)	430
He-dibromomethane	Fuller et al. (1969)	428
He-1-iodobutane	Fuller et al. (1969)	428
He-2-iodobutane	Fuller et al. (1969)	427
He-hexafluorobenzene	Fuller et al. (1969)	429
He-CF ₃ I	Belousova et al. (1970)	300
He-C ₃ F ₇ I	Belousova et al. (1970)	300
He-UF ₆	Ljunggren (1965)	293
He-As ₄	Krol et al. (1967)	733 to 913
Ne-H ₂	Paul and Srivastava (1961)	242 to 341
	Weissman and Mason (1962 b)	90 to 523
Ne-D ₂	Weissman and Mason (1962 b)	293
Ne-NH ₃	Srivastava (1962)	274 to 333
Ne-CD ₃ H	Vugts et al. (1971)	233 to 422
Ne-Na	Anderson and Ramsey (1963)	425
	Violino (1968)	425
Ne-N ₂	DiPippo et al. (1967)	293 and 303
Ne-CO ₂	Weissman et al. (1961)	242 to 427
	Breetveld et al. (1966, 1967)	293 and 303
	Annis et al. (1969)	295
Ne-Rb	Franzen (1959)	320
	Violino (1968)	340
Ne-Cs	Legowski (1964)	299
	Violino (1968)	317
Ne-CF ₃ I	Belousova et al. (1970)	300
Ne-Hg	Tubbs (1967)	323-333
Ne-C ₃ F ₇ I	Belousova et al. (1970)	300
Ne-UF ₆	Ljunggren (1965)	293
Ar-H ₂	Waldmann (1944, 1947)	293
	Strehlow (1953)	288 to 418
	Paul and Srivastava (1961 c)	242 to 341
	Suetin and Ivakin (1961)	291
	Westenberg and Frazier (1962)	295 to 1069
	Weissman and Mason (1962 b)	293 to 523
	Golubev and Bondarenko (1963)	298 to 363
	Ivakin and Suetin (1964 b)	291 to 473
	Mason et al. (1964 a)	294
	Suetin (1964)	291
	Cordes and Kerl (1965)	296
	Evans and Kenney (1965)	293
	Kosov and Kurlapov (1966)	295
	Arnold and Toor (1967)	307
	Mason et al. (1967)	296
	Annis et al. (1969)	295
Ar-D ₂	Ivakin and Suetin (1964 a)	297
Ar-T ₂	Mason et al. (1964 a)	295
	Annis et al. (1969)	295
Ar-CH ₄	Carswell (1960)	298
	Carswell and Stryland (1963)	298
	Arnold and Toor (1967)	307
	Jacobs et al. (1970)	298
Ar-NH ₃	Ivakin and Suetin (1964 a)	295
	Srivastava and Srivastava (1962)	255 to 333
	DiPippo et al. (1967)	293 and 303
Ar-H ₂ O	O'Connell et al. (1969)	282 to 353
Ar-CD ₃ H	Vugts et al. (1971)	233 to 422

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

b. Noble Gases and Another Component—Continued		
System	Reference	T, K
Ar-Na	von Hartel et al. (1932)	654
Ar-C ₂ H ₂	Violino (1968)	(?)
	Suetin and Ivakin (1961)	287
Ar-C ₂ H ₄	Suetin (1964)	287
	Ivakin et al. (1968)	298 to 407
Ar-N ₂	Weissman (1964)	298
	Waldmann (1944, 1947)	293
Ar-CO	Schäfer and Moesta (1954)	233 to 363
	Westenberg and Walker (1957)	293
Ar-C ₂ H ₆	Paul and Srivastava (1961 b)	244 to 335
	DiPippo et al. (1967)	293 and 303
Ar-O ₂	Ivakin and Suetin (1964 a)	296
	Jacobs et al. (1970)	298
Ar-air	Waldmann (1944, 1947)	293
	Paul and Srivastava (1961 a)	243 to 334
Ar-K	Scott and Dullien (1962)	293
	Evans et al. (1969)	295
Ar-C ₃ H ₈	Khomchenkov et al. (1968)	723
	Ivanovskii et al. (1969)	630 to 950
Ar-CO ₂	Jacobs et al. (1970)	298
	Lannus and Grossman (1970 a, b)	242 to 473
Ar-Fe	Waldmann (1944, 1947)	293
	Suetin and Ivakin (1961)	289
Ar- <i>n</i> -butane	Ivakin and Suetin (1964 b)	289 to 473
	Holsen and Strunk (1964)	276 and 317
Ar-Cr	Suetin (1964)	289
	Pakurar and Ferron (1964)	295, and 1181 to 1676
Ar-nitromethane	Kestin et al. (1966)	293 and 303
	Kosov and Novosad (1966 a)	294
Ar-SO ₂	Pakurar and Ferron (1966)	1132 to 1798
	Ferron (1967)	1100 to 1800
Ar-Zn	Oost et al. (1967)	295 and 343
	Gurvich and Matizen (1968)	308
Ar- <i>n</i> -pentane	Grievesson and Turkdogan (1964)	1600
	Grievesson and Turkdogan (1964)	1600
Ar-ether	Hargrove and Sawyer (1967)	298 to 473
	Hargrove and Sawyer (1967)	298 to 473
Ar-benzene	Hargrove and Sawyer (1967)	298 to 473
	Jacobs et al. (1970)	298
Ar-HBr	Grievesson and Turkdogan (1964)	1600
	Grievesson and Turkdogan (1964)	1600
Ar-cyclohexane	Byrne et al. (1967)	303
	Schäfer (1959)	263
Ar-methylcyclopentane	Nikolaev and Aleskovskii (1964)	1100 to 2600
	Hargrove and Sawyer (1967)	298 to 473
Ar- <i>n</i> -hexane	Hargrove and Sawyer (1967)	298 to 473
	Hargrove and Sawyer (1967)	298 to 473
Ar-2,3-dimethylbutane	Hargrove and Sawyer (1967)	298 to 473
	Cummings and Ubbelohde (1953, 1955)	289
Ar-3-pentanone	Barr and Sawyer (1964)	300
	Fairbanks and Wilke (1950)	294
Ar-toluene	Byrne et al. (1967)	303
	Clarke and Ubbelohde (1957)	303
Ar-monofluorobenzene	Clarke and Ubbelohde (1957)	303
	Cummings and Ubbelohde (1953, 1955)	303
Ar-2,4-dimethylpentene	Clarke and Ubbelohde (1957)	303
	Cummings and Ubbelohde (1953, 1955)	303
Ar- <i>n</i> -heptane	Clarke and Ubbelohde (1957)	303
	Legowski (1964)	299
Ar-2,2,4-trimethylpentane	Khomchenkov et al. (1968)	723
	Violino (1968)	299 and 317
Ar-Cs	Suetin and Ivakin (1961)	287
	Suetin (1964)	287
Ar-SF ₆	Ivakin and Suetin (1964 b)	287 to 472
	Evans and Kenney (1965)	293
Ar-1-bromo-3-methylbutane	Byrne et al. (1967)	303
	Mackenzie and Melville (1933)	289
Ar-Br ₂	Belousova et al. (1970)	300
	Spencer et al. (1969)	459 to 607
Ar-CF ₃ I	Belousova et al. (1970)	300
	Ljunggren (1965)	293
Ar-UF ₆	Krol et al. (1967)	853 to 913
	Miller and Carman (1961)	293
Ar-As ₄	Miller and Carman (1961)	293
	Mason et al. (1964 b)	296

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

b. Noble Gases and Another Component—Continued		
System	Reference	<i>T</i> , K
Kr-D ₂	Miller and Carman (1964)	293
	Fedorov et al. (1966)	291
	Annis et al. (1968)	77 to 760
	Annis et al. (1969)	295
	Mason et al. (1964 b)	255 to 362
Kr-T ₂	Annis et al. (1969)	295
	Mason et al. (1964 b)	252 to 346
Kr-NH ₃	Annis et al. (1969)	295
Kr-NH ₃	Srivastava and Srivastava (1962)	255 to 333
Kr-C ₂ H ₄	Durbin and Kobavashi (1962)	298
Kr-N ₂	Durbin and Kobayashi (1962)	248 and 308
Kr-CO	Singh et al. (1967)	274 to 319
Kr-NO	Singh et al. (1967)	274 to 318
Kr-O ₂	Ivakin et al. (1967)	298 to 408
Kr-air	Reist (1967)	273
Kr-CO ₂	Durbin and Kobayashi (1962)	308
Kr-acetone	Kestin and Yata (1968)	293 and 303
	Srivastava and Saran (1966 a)	284 to 313
Kr-SO ₂	Saran and Singh (1966)	303
Kr-C ₂ H ₅ Cl	Srivastava and Saran (1966 b)	274 to 318
	Singh and Srivastava (1968)	275 to 318
Kr-(C ₂ H ₅) ₂ O	Srivastava and Saran (1966 b)	274 to 318
Kr-CH ₂ Cl ₂	Singh and Srivastava (1968)	278 to 318
Kr-Rb	Franzen (1959)	320
	Violino (1968)	320
Kr-CHCl ₃	Srivastava and Saran (1966 a)	284 to 313
Kr-Hg	Nakayama (1968)	301
Kr-UF ₆	Ljunggren (1965)	293
Xe-H ₂	Paul and Srivastava (1961 c)	242 to 341
	Weissman and Mason (1962 b)	293 to 550
Xe-NH ₃	Miller and Carman (1964)	293
	Srivastava (1962)	274 to 331
Xe-N ₂	Paul and Srivastava (1961 b)	242 to 334
Xe-O ₂	Paul and Srivastava (1961 a)	242 to 334
Xe-Rb	Franzen (1959)	320
	Violino (1968)	320
Xe-CF ₃ I	Belousova et al. (1970)	300
Xe-Hg	Nakayama (1968)	301
Xe-C ₃ F ₇ I	Belousova et al. (1970)	300
Xe-UF ₆	Ljunggren (1965)	293
The list of studies for Rn mixtures is not comprehensive, and references to other studies are given by Hirst and Harrison (1939) and by Raabe (1968).		
Rn-H ₂	Hirst and Harrison (1939)	288
Rn-air	Rutherford and Brooks (1901)	Room temperature
	Hirst and Harrison (1939)	288
	Korpusov et al. (1964)	(?)
	Vučić and Milojević (1966)	(?)
	Raabe (1968)	299

c. Dissociated Gases

System	Reference	<i>T</i> , K
H-He	Khouw et al. (1969)	275
H-Ar	Wise (1959)	293 (assumed)
	Khouw et al. (1969)	275
H-H ₂	Wise (1959)	293 (assumed)
	Wise (1961)	293 to 719
	Weissman and Mason (1962 a)	200 to 1000
	Browning and Fox (1964)	190 to 373
	Khouw et al. (1969)	202 to 364
N-N ₂	Sancier and Wise (1969)	293 to 719
	Young (1961)	298 (assumed)
	Morgan and Schiff (1964)	280
O-He	Morgan and Schiff (1964)	280
	Yolles and Wise (1968)	298
O-Ar	Morgan and Schiff (1964)	280
	Yolles and Wise (1968)	298
	Baker (1970 b)	298
O-Kr	Yolles and Wise (1968)	298
O-N ₂	Morgan and Schiff (1964)	280
O-O ₂	Krongelb and Strandberg (1959)	300
	Walker (1961)	298
	Morgan and Schiff (1964)	280
O-O ₂	Yolles and Wise (1968)	298
	Yolles et al. (1970)	298 to 873

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures			
System	Reference	T, K	
H ₂ -H ₂	Harteck and Schmidt (1933)	20 to 293	
	Lipsicas (1962)	56 to 90	
	Hartland and Lipsicas (1963)	20	
	Amdur and Beatty (1965)	195 to 353	
	Mason et al. (1965)	295	
	Reichenbacher et al. (1965)	297	
H ₂ -HD	Annis et al. (1969)	295	
	Weissman and Mason (1962 b)	72 to 293	
H ₂ -TH	Reichenbacher et al. (1965)	297	
	Amdur and Beatty (1965)	195 and 273	
H ₂ -D ₂	Heath et al. (1941)	288	
	Waldmann (1944, 1947)	293	
	Bendt (1958)	14 to 296	
	McCarty and Mason (1960)	303	
	Weissman and Mason (1962 b)	72 to 293	
	Ivakin and Suetin (1964 a)	296	
H ₂ -DT	Diller and Mason (1966)	14 to 293	
	Reichenbacher et al. (1965)	297	
	Amdur and Beatty (1965)	195 to 353	
H ₂ -T ₂	Mason et al. (1965)	295	
	Reichenbacher et al. (1965)	297	
H ₂ -CH ₄	von Obermayer (1883)	273 to 289	
	Boyd et al. (1951)	298	
	Fejes and Czárán (1961)	298	
	Weissman and Mason (1962 b)	293 to 523	
	Arnold and Toor (1967)	307	
	Mason et al. (1967)	296	
H ₂ -NH ₃	Bunde (1955)	298 to 358	
	Schäfer (1959)	240 to 403	
	Scott and Cox (1960)	273 to 533	
	Ivakin and Suetin (1964 a)	297	
	Weissman (1964)	293 to 523	
	Pal and Barua (1967)	306 to 479	
H ₂ -H ₂ O	Guglielmo (1882)	291	
	Winkelmann (1884 a, b)	323 and 366	
	Winkelmann (1889)	293 and 372	
	Mache (1910)	300 to 366	
	Trautz and Müller (1935)	293 to 372	
	McMurtie and Keyes (1948)	303 to 333	
	Hippenmeyer (1949)	283 to 368	
	Schwartz and Brow (1951)	307 to 353	
	Crider (1956)	307 and 329	
	Nelson (1956)	298 to 328	
	H ₂ -Na	von Hartel and Polanyi (1930)	633
		von Hartel et al. (1932)	655
		Ramsey and Anderson (1964)	473
H ₂ -C ₂ H ₂	Violino (1968)	473	
	Weissmann (1964)	293 to 373	
H ₂ -N ₂	Lonius (1909)	285-287	
	Boardman and Wild (1937)	288	
	Waldmann (1944, 1947)	293	
	Schäfer et al. (1951)	193 to 336	
	Nettley (1954)	288	
	Schäfer and Moesta (1954)	200 to 400	
	Bunde (1955)	298 to 358	
	van Itterbeek and Nihoul (1957)	137 and 153	
	Weisz (1957)	293 (?)	
	Vyshenskaya and Kosov (1959)	293 to 1083	
	Giddings and Seager (1960)	293	
	Scott and Cox (1960)	294 to 573	
	Bohemen and Purnell (1961)	324	
	Fejes and Czárán (1961)	298	
	Suetin and Ivakin (1961)	289	
	Giddings and Seager (1962)	297	
	Scott and Dullien (1962)	293	
	Weissman and Mason (1962 b)	82 to 523	
	Bondarenko and Golubev (1964)	273 to 473	
	Ivakin and Suetin (1964 b)	289 to 471	
	Suetin (1964)	289	
	Cordes and Kerl (1965)	296	
	Vyshenskaya and Kosov (1965)	293 to 1083	
	van Heijningen et al. (1966)	65 to 295	
	Pal and Barua (1967)	307 to 478	
	Kestin and Yata (1968)	293 and 303	
	Schnieder and Schäfer (1969)	273 to 1300	
	Saxena and Gupta (1970)	313 to 366	
	H ₂ -CO	Loschmidt (1870 b)	293
		von Obermayer (1883)	282-285
		Weissman and Mason (1962 b)	195 to 523
		Ivakin and Suetin (1964 a)	296

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued			
System	Reference	<i>T</i> , K	
$H_2-C_2H_4$	Ivakin and Suetin (1964 b)	296 to 471	
	von Obermayer (1883)	287	
H_2-NO	Weissman (1964)	195 to 523	
	Weissman (1964)	273	
$H_2-C_2H_6$	von Obermayer (1883)	288	
	Boyd et al. (1951)	298	
H_2-O_2	Fejes and Czárán (1961)	298	
	Weissman (1964)	293 to 523	
	Loschmidt (1870 a)	252 to 286	
	Loschmidt (1870 b)	252 to 289	
	Wretschko (1870)	294 and 297	
	von Obermayer (1880)	286 and 335	
	von Obermayer (1883)	281 to 291	
	Lonius (1909)	284 to 288	
	van Itterbeek and Nihoul (1957)	142 and 153	
	Walker and Westenberg (1960)	295 to 900	
	Weissman and Mason (1962 b)	294 to 550	
	Walker and Westenberg (1966)	295 to 901	
	Zhargasov and Kosov (1968)	103 to 298	
H_2 -air	Saxena and Gupta (1970)	313 to 366	
	von Obermayer (1883)	281 to 284	
	Barus (1924 b)	297	
	Kosov (1957)	289	
	Suetin et al. (1960)	273	
	Currie (1960)	285 to 309	
	Suetin and Ivakin (1961)	292	
	Ivakin and Suetin (1964 a)	297	
	Suetin (1964)	292	
	Evans et al. (1969)	295	
	H_2-CH_3OH	Winkelmann (1885)	299 and 323
		Huang et al. (1968)	353 to 423
	H_2-HCl	Weissman (1964)	293 to 523
H_2-K	Ivanovskii et al. (1969)	680 to 830	
$H_2-C_3H_8$	Weissman (1964)	293 to 523	
H_2-CO_2	Loschmidt (1870 a)	273 and 286	
	Loschmidt (1870 b)	273 to 289	
	Wretschko (1870)	297	
	von Obermayer (1880)	285 and 335	
	von Obermayer (1882 a)	284 to 293	
	von Obermayer (1883)	280 to 294	
	Schmidt (1904)	288	
	Lonius (1909)	286 to 294	
	Boardman and Wild (1937)	288	
	Waldmann (1944, 1947)	293	
	Boyd et al. (1951)	298	
	Schäfer et al. (1951)	252 to 308	
	Lonsdale and Mason (1957)	259 to 358	
	Saxena and Mason (1959)	250 to 368	
	Vyshenskaya and Kosov (1959)	293 to 1083	
	McCarty and Mason (1960)	303	
	Suetin et al. (1960)	273	
	Miller and Carman (1961)	293	
	Suetin and Ivakin (1961)	292	
	Wicke and Hugo (1961)	295	
	Giddings and Seager (1962)	300	
	Bondarenko and Golubev (1964)	323 and 363	
	Ivakin and Suetin (1964 b)	292 to 473	
	Miller and Carman (1964)	293	
	Suetin (1964)	292	
	Weissman (1964)	298 to 550	
	Vyshenskaya and Kosov (1965)	293 to 1083	
	Mason et al. (1967)	296	
	Annis et al. (1969)	295	
	Schneider and Schäfer (1969)	273 to 990	
	Kosov and Zhargasov (1970)	196 to 298	
	H_2-N_2O	von Obermayer (1883)	283
		Weissman (1964)	300 to 550
$H_2-C_3H_8$	Fejes and Czárán (1961)	298	
H_2 -formic acid	Weissman (1964)	273 to 550	
	Winkelmann (1885)	339 and 358	
$H_2-C_2H_5OH$	Baumgartner (1877 a)	291	
	Winkelmann (1884 a)	314 and 340	
	Winkelmann (1885)	323 and 337	
	Trautz and Müller (1935)	340	
	Huang et al. (1968)	353 to 453	
H_2 -2-butene	Weissman (1964)	293 to 523	
H_2 -acetone	Trautz and Müller (1935)	296	
H_2 - <i>n</i> -butane	Strehlow (1953)	288 to 430	
H_2 -acetic acid	Fejes and Czárán (1961)	298	
	Winkelmann (1885)	339 to 372	

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	T, K
H ₂ - <i>n</i> -propyl alcohol	Winkelmann (1885)	340 and 357
H ₂ -nitromethane	Byrne et al. (1967)	303
H ₂ -SO ₂	Loschmidt (1870 b)	286
	Schäfer (1959)	263 to 473
	Weissman (1964)	290 to 472
H ₂ - <i>n</i> -pentane	Huang et al. (1968)	353 to 453
H ₂ -ethyl formate	Winkelmann (1884 c)	294 and 319
H ₂ -methyl acetate	Winkelmann (1884 c)	294 and 319
H ₂ -propionic acid	Winkelmann (1885)	366 and 372
H ₂ - <i>n</i> -butyl alcohol	Winkelmann (1885)	372
	Huang et al. (1968)	393 to 483
H ₂ - <i>i</i> -butyl alcohol	Winkelmann (1885)	340 and 357
H ₂ - <i>sec</i> -butyl alcohol	Huang et al. (1968)	393 to 483
H ₂ -ethyl ether	Stefan (1873)	292
	Baumgartner (1877 a)	290
	Winkelmann (1884 a)	284 and 293
	Trautz and Müller (1935)	273 and 293
	Weissman (1964)	288 to 486
H ₂ -CS ₂	Baumgartner (1877 a)	290
	Baumgartner (1877 b)	268 to 311
H ₂ -benzene	Trautz and Ludwig (1930)	296
	Trautz and Ries (1931)	296
	Trautz and Müller (1935)	296
	Hudson et al. (1960)	311
	Huang et al. (1968)	373 to 483
H ₂ -pyridine	Hudson et al. (1960)	318
H ₂ -2:3-dimethylbuta-1:3-diene	Cummings et al. (1955)	288
H ₂ -hexa-1:5-diene	Cummings et al. (1955)	288
H ₂ -thiophen	Hudson et al. (1960)	302
H ₂ -cyclohexane	Cummings and Ubbelohde (1953, 1955)	289
	Hudson et al. (1960)	289
	Huang et al. (1968)	373 to 453
H ₂ -2:3-dimethylbut-2-ene	Cummings et al. (1955)	288
H ₂ -methyl cyclopentane	Cummings and Ubbelohde (1953, 1955)	289
H ₂ -piperidine	Hudson et al. (1960)	315
H ₂ -Rb	McNeal (1962)	343
	Violino (1968)	343
H ₂ -2:3-dimethylbutane	Cummings and Ubbelohde (1953, 1955)	289
H ₂ - <i>n</i> -hexane	Cummings and Ubbelohde (1953, 1955)	289
	Huang et al. (1968)	353 to 453
H ₂ - <i>n</i> -butyric acid	Winkelmann (1885)	372
H ₂ - <i>i</i> -butyric acid	Winkelmann (1885)	371
H ₂ -ethyl acetate	Winkelmann (1884 c)	319
H ₂ -methyl propionate	Winkelmann (1884 c)	319 and 340
H ₂ -propyl formate	Winkelmann (1884 c)	319 and 340
H ₂ - <i>n</i> -amyl alcohol	Winkelmann (1885)	372
H ₂ -active amyl alcohol	Winkelmann (1885)	372
H ₂ -tetrahydrothiophen	Hudson et al. (1960)	318
H ₂ -toluene	Fairbanks and Wilke (1950)	301
	Huang et al. (1968)	393 to 483
H ₂ -monofluorobenzene	Byrne et al. (1967)	303
H ₂ - <i>n</i> -heptane	Cummings et al. (1955)	303
	Clarke and Ubbelohde (1957)	303
H ₂ -2:4-dimethylpentane	Clarke and Ubbelohde (1957)	303
H ₂ -triethylamine	Mehta (1966)	298
H ₂ -ethyl propionate	Winkelmann (1884 c)	340 and 363
	Fairbanks and Wilke (1950)	301
H ₂ -methyl butyrate	Winkelmann (1884 c)	340 and 365
H ₂ -methyl <i>i</i> -butyrate	Winkelmann (1884 c)	323 and 340
H ₂ - <i>i</i> -valeric acid	Winkelmann (1885)	372
H ₂ - <i>n</i> -hexyl alcohol	Winkelmann (1885)	372
H ₂ - <i>n</i> -octane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
H ₂ -2:2:4-trimethylpentane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
H ₂ - <i>i</i> -butyl acetate	Winkelmann (1884 c)	340 and 371
H ₂ -ethyl butyrate	Winkelmann (1884 c)	340 and 370
H ₂ -ethyl <i>i</i> -butyrate	Winkelmann (1884 c)	340 and 369
H ₂ -propyl propionate	Winkelmann (1884 c)	370
H ₂ -CHCl ₃	Baumgartner (1877 a)	291
H ₂ -CF ₂ Cl ₂	Miller and Carman (1961, 1964)	293
H ₂ - <i>n</i> -nonane	Cummings et al. (1955)	340
H ₂ - <i>i</i> -butyl propionate	Winkelmann (1884 c)	371
H ₂ -propyl butyrate	Winkelmann (1884 c)	371
H ₂ -propyl <i>i</i> -butyrate	Winkelmann (1884 c)	370
H ₂ -ethyl valerate	Winkelmann (1884 c)	371
H ₂ -1-bromobutane	Byrne et al. (1967)	303
H ₂ -2:3:3-trimethylheptane	Cummings and Ubbelohde (1953, 1955)	364
H ₂ - <i>n</i> -decane	Cummings and Ubbelohde (1953, 1955)	364
H ₂ -amyl propionate	Winkelmann (1884 c)	371

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	<i>T</i> , K
H ₂ - <i>i</i> -butyl butyrate	Winkelmann (1884 c)	371
H ₂ - <i>i</i> -butyl <i>i</i> -butyrate	Winkelmann (1884 c)	371
H ₂ -propyl valerate	Winkelmann (1884 c)	371
H ₂ -SF ₆	Boyd et al. (1951)	298
	Strehlow (1953)	286 to 418
	Suetin and Ivakin (1961)	290
	Ivakin and Suetin (1964 b)	290 to 473
	Suetin (1964)	290
	Evans and Kenney (1965)	293
H ₂ -1-bromo-3-methylbutane	Byrne et al. (1967)	303
H ₂ -CCl ₄	Trautz and Ries (1931)	296
	Trautz and Müller (1935)	296
H ₂ -amyl <i>i</i> -butyrate	Winkelmann (1884 c)	371
H ₂ - <i>i</i> -butyl valerate	Winkelmann (1884 c)	371
H ₂ -Br ₂	Mackenzie and Melville (1932)	286
	Mackenzie and Melville (1933)	290
H ₂ -1-iodopropane	Byrne et al. (1967)	303
H ₂ - <i>n</i> -dodecane	Cummings and Ubbelohde (1953, 1955)	400
H ₂ -Hg	Gaede (1915)	Room temperature
	Spier (1939)	314 to 325
H ₂ -di- <i>n</i> -butyl phthalate	Birks and Bradley (1949)	293
H ₂ -UF ₆	Ljunggren (1965)	293
HD-D ₂	Weissman and Mason (1962 b)	72 to 293
D ₂ -TH	Reichenbacher et al. (1965)	297
	Amdur and Beatty (1965)	195 and 273
D ₂ -DT	Reichenbacher et al. (1965)	297
D ₂ -T ₂	Amdur and Beatty (1965)	195 to 353
	Mason et al. (1965)	295
	Reichenbacher et al. (1965)	297
D ₂ -NH ₃	Ivakin and Suetin (1964 a)	297
D ₂ -N ₂	Ivakin and Suetin (1964 a)	297
	Saxena and Gupta (1970)	313 to 366
D ₂ -CO	Ivakin and Suetin (1964 a)	296
D ₂ -air	Ivakin and Suetin (1964 a)	297
D ₂ -CO ₂	Saxena and Mason (1959)	250 to 372
	Ivakin and Suetin (1964 a)	296
	Annis et al. (1969)	295
D ₂ - <i>n</i> -heptane	Clarke and Ubbelohde (1957)	303
D ₂ -2:4-dimethylpentane	Clarke and Ubbelohde (1957)	303
D ₂ - <i>n</i> -octane	Clarke and Ubbelohde (1957)	303
D ₂ -2:2:4-trimethylpentane	Clarke and Ubbelohde (1957)	303
D ₂ -SF ₆	Ivakin and Suetin (1964 a)	296
T ₂ -N ₂	Mason et al. (1964 a)	297
	Annis et al. (1969)	295
T ₂ -CO ₂	Mason et al. (1964 a)	298
	Annis et al. (1969)	295
CH ₄ -CH ₄	Winn and Ney (1947)	293
	Winn (1950)	90 to 353
	Ember et al. (1964)	297
	Mueller and Cahill (1964)	298 to 383
	Hu and Kobayashi (1970)	298
	Mistler et al. (1970)	293
	Weissman (1964)	288
CH ₄ -NH ₃	Schwartz and Brow (1951)	308 to 352
CH ₄ -H ₂ O	Kimpton and Wall (1952)	298 to 333
	O'Connell et al. (1969)	283 to 328
	Table 1 has a misprint, the highest temperature is not 323 K but 328 K.	
CH ₄ -CH ₃ T	Hu and Kobayashi (1970)	298
CH ₄ -C ₂ H ₂	Weissman (1964)	313 to 373
CH ₄ -N ₂	Mueller and Cahill (1964)	298 to 383
	Arai et al. (1967)	313 and 373
	Jacobs et al. (1970)	298
CH ₄ -CO	Mueller and Cahill (1964)	298 to 383
CH ₄ -C ₂ H ₆	Weissman (1964)	293 to 523
	Chang (1966)	255 to 311
	Gover (1967)	295
CH ₄ -O ₂	Walker and Westenberg (1960, 1966, 1968)	294 to 985
CH ₄ -air	Coward and Georgeson (1937)	289 to 295
CH ₄ -CO ₂	von Obermayer (1887)	294–295
	Fejes and Czárán (1961)	298
	Ember et al. (1964)	297
	Weissman (1964)	298
	Kestin and Yata (1968)	293 and 303
	Hawtin et al. (1969)	293 to 873
	Weissman (1969)	293 to 370
CH ₄ -C ₃ H ₈	Weissman (1964)	293 to 523
	Chang (1966)	255 to 311
CH ₄ -CH ₃ Cl	Manner (1967)	298 to 478

TABLE 16. Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued

d. Other Mixtures—Continued		
System	Reference	T, K
CH ₄ -n-C ₄ H ₁₀	Chang (1966)	255 to 311
	Kestin and Yata (1968)	293 and 303
CH ₄ -Rb	Beatty (1969)	303
	McNeal (1962)	333
	Violino (1968)	333
CH ₄ -n-hexane	Carmichael et al. (1955 b)	294 to 377
	Kohn and Romero (1965)	298 to 333
CH ₄ -3-methylpentane	Kohn and Romero (1965)	298 to 333
CH ₄ -CF ₄	Mueller and Cahill (1964)	298 to 383
	Hu and Kobayashi (1970)	298
CH ₄ -n-heptane	Carmichael et al. (1955 a)	311 to 377
	Reamer and Sage (1963)	311 to 377
CH ₄ -SF ₆	Manner (1967)	298 to 478
CH ₄ -Br ₂	Mackenzie and Melville (1933)	289
CH ₄ -T-CF ₄	Hu and Kobayashi (1970)	298
CD ₄ -H-CD ₄	Vugts et al. (1971)	233 to 422
NH ₃ -NH ₃	Paul and Watson (1966)	233 to 353
	Baker (1970 a)	301 to 446
NH ₃ -N ₂	Bunde (1955)	298 to 358
	Ivakin and Suetin (1964 a)	295
	Weissman (1964)	293 to 523
NH ₃ -CO	Ivakin and Suetin (1964 a)	295
NH ₃ -C ₂ H ₄	Weissman (1964)	293 to 523
NH ₃ -air	Toepler (1896)	292
	Wintergerst (1930)	287 to 298
	Andrew (1955)	293
	Ivakin and Suetin (1964 a)	295
	Burch and Raw (1967)	273 to 673
	Weissman (1964)	293 to 473
	Srivastava and Srivastava (1963)	288 to 338
	Pal and Bhattacharyya (1969)	299 to 373
	Ivakin and Suetin (1964 a)	297
	Ferron (1967)	950 to 1400
H ₂ O-H ₂ O	Hippenmeyer (1949)	273 to 368
	Schwartz and Brow (1951)	307 to 352
	Bose and Chakraborty (1955-56)	332 and 336
	Crider (1956)	329 and 349
	Nelson (1956)	298 to 328
	O'Connell et al. (1969)	282 to 373
	Nagata and Hasegawa (1970)	393 to 423
	Schwartz and Brow (1951)	308 to 353
	Kimpton and Wall (1952)	298
	Kimpton and Wall (1952)	298
H ₂ O-C ₂ H ₆	Bose and Chakraborty (1955-56)	332 and 336
	Schwartz and Brow (1951)	308 to 352
H ₂ O-O ₂	Walker and Westenberg (1960, 1966)	390 to 1070
	Stefan (1871)	Room temperature
H ₂ O-air	Guglielmo (1881, 1882)	280 to 296
	Winkelmann (1884 a, b)	323 and 366
	Winkelmann (1888)	290 to 294
	Winkelmann (1889)	290 and 372
	Houdaille (1896)	273
	Brown and Escombe (1900)	286 to 290
	Mache (1910)	301 to 366
	Le Blanc and Wuppermann (1916)	315
	Summerhays (1930)	289-290
	Houghton (1933)	293
	Ackermann (1934)	356 to 366
	Gilliland (1934)	299 to 332
	Trautz and Müller (1935)	281 to 373
	Schirmer (1938)	273 to 370
	Klibanova et al. (1942)	373 to 1493
	Brookfield et al. (1947)	298 to 318
	Kimpton and Wall (1952)	298
	Rossié (1953)	356 to 575
	Lee and Wilke (1954)	298
	Bose and Chakraborty (1955-56)	327 and 336
Narsimhan (1955-56)	303	
Nelson (1956)	298	
Richardson (1959)	319	
Petit (1965)	293 to 301	
Weissman (1968 a)	373	
H ₂ O-H ₂ O ₂	Weissman (1968 a)	443 to 513
	Guglielmo (1882)	291
H ₂ O-CO ₂	Winkelmann (1884 a, b)	323 and 366
	Winkelmann (1889)	294-298 and 373
H ₂ O-CO ₂	Trautz and Müller (1935)	294 to 372
	Schwartz and Brow (1951)	307 to 352
	Rossié (1953)	433
	Crider (1956)	329 and 349

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	T, K
	Ember et al. (1964)	1000 to 1400
	Ferron (1967)	1000 to 1700
	Nagata and Hasegawa (1970)	394 to 423
H ₂ O-C ₃ H ₈	Kimpton and Wall (1952)	298
H ₂ O-C ₂ H ₅ OH	Weissman (1968 a)	373
H ₂ O-SO ₂	Kimpton and Wall (1952)	298
H ₂ O-ethyl ether	Winkelmann (1884 a)	284 and 293
H ₂ O-CCl ₂ F ₂	Lee and Wilke (1954)	298
D ₂ O-air	Kimpton and Wall (1952)	298 to 333
Na-N ₂	von Hartel and Polyani (1930)	633
	von Hartel et al. (1932)	655
	Van der Held and Miesowicz (1937)	288
	Cvetanović and Le Roy (1952)	527
	Ramsey and Anderson (1964)	453
	Violino (1968)	453
	von Hartel et al. (1932)	655
Na-C ₃ H ₁₂	Mueller and Cahill (1964)	298 to 383
C ₂ H ₂ -C ₂ H ₂	Mueller and Cahill (1964)	298 to 383
C ₂ H ₂ -C ₂ H ₄	Mueller and Cahill (1964)	298 to 383
C ₂ H ₂ -C ₂ H ₆	Mueller and Cahill (1964)	298 to 383
C ₂ H ₂ -O ₂	Suetin and Ivakin (1961)	287
	Suetin (1964)	287
C ₂ H ₂ -air	Kosov (1957)	289
	Suetin and Ivakin (1961)	288
	Suetin (1964)	288
C ₂ H ₂ -C ₃ H ₆	Weissman (1964)	313 to 373
HCN-air	Klotz and Miller (1947)	273
N ₂ -N ₂	Winn (1948)	293
	Winn (1950)	78 to 353
	Winter (1951)	273 and 318
	DeLuca (1954)	273
	Amdur and Mason (1958)	1000 to 15 000
	Belyaev and Leonas (1966)	2000 to 15 000
	Vugts et al. (1970)	233 to 422
N ₂ -CO	Boardman and Wild (1937)	288-291
	Wicke and Hugo (1961)	295
	Weissman and Mason (1962 b)	300 to 550
	Amdur and Shuler (1963)	195 to 373
	Ivakin and Suetin (1964 a)	296
	Vugts et al. (1970)	233 to 422
N ₂ -C ₂ H ₄	Boyd et al. (1951)	298
	Knox and McLaren (1963, 1964)	291
	Mueller and Cahill (1964)	298 and 373
	Weissman (1964)	300 to 550
	Evans and Kenney (1965)	287 and 291
	Fuller and Giddings (1967)	303
	Fuller et al. (1969)	303
N ₂ -NO	Weissman (1964)	293 and 373
N ₂ -C ₂ H ₆	Boyd et al. (1951)	298
	Fejes and Czárán (1961)	298
	Arai et al. (1967)	313 and 373
	Jacobs et al. (1970)	298
N ₂ -O ₂	von Obermayer (1880)	286-287 and 335
	von Obermayer (1882 b)	286-289
	Loniuss (1909)	285-286
	Parker and Hottel (1936)	1157
	Waldmann (1944, 1947)	293
	Bohemen and Purnell (1961)	324
	Giddings and Seager (1962)	298
	Weissman and Mason (1962 b)	300 to 550
	Belyaev and Leonas (1966)	2000 to 15 000
	Arnikar et al. (1967 a, b)	298
	Saxena and Gupta (1970)	313 to 366
N ₂ -CH ₃ OH	Arnikar and Ghule (1969)	355
N ₂ -HCl	Mian (1967)	324 to 523
	Mian et al. (1969)	324 to 523
N ₂ -K	Ivanovskii et al. (1969)	630 to 920
N ₂ -CO ₂	Parker and Hottel (1936)	1157
	Boardman and Wild (1937)	288-290
	Wicke and Kallenbach (1941)	273
	Waldmann (1944, 1947)	293
	Boyd et al. (1951)	298
	Schäfer et al. (1951)	252 to 308
	Westenberg and Walker (1957)	293
	Walker (1958)	296 to 1114
	Walker and Westenberg (1958 a)	298 to 1150
	Walker and Westenberg (1958 b)	300 to 1150
	Vyshenskaya and Kosov (1959)	293 to 1083
	Walker et al. (1960)	299
	Bohemen and Purnell (1961)	324
	Suetin and Ivakin (1961)	290

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	T, K
	Wicke and Hugo (1961)	295
	Giddings and Seager (1962)	293 to 299
	Ivakin and Suetin (1964 b)	290 to 473
	Suetin (1964)	290
	Pakurar and Ferron (1964)	295, 1156 to 1653
	Weissman (1964)	293
	Vyshenskaya and Kosov (1965)	293 to 1083
	Kestin et al. (1966)	293 and 304
	Pakurar and Ferron (1966)	1081 to 1810
	Walker and Westenberg (1966,	296 to 1114
	Coates and Mian (1967)	301 to 525
	Ferron (1967)	1100 to 1800
	Mian (1967)	301 to 525
	Ellis and Holsen (1969)	298 to 880
	Schneider and Schäfer (1969)	273 to 1300
	Humphreys and Gray (1970)	300 to 1800
	Lannus and Grossmann (1970 a, b)	283 to 399
	Nagata and Hasegawa (1970)	314 to 365
$N_2-C_3H_8$	Fejes and Czárán (1961)	298
	Arai et al. (1967)	313 and 373
	Jacobs et al. (1970)	298
$N_2-C_2H_5OH$	Lannus and Grossmann (1970 a, b)	283 to 472
	Bose and Chakraborty (1955-56)	327 and 331
	Arnikar et al. (1967 b)	353
	Arnikar and Ghule (1969)	355
N_2 -acetone	Mehta (1966)	290
	Arnikar et al. (1967 b)	353
	Arnikar and Ghule (1969)	353
N_2 - <i>n</i> -butane	Nagata and Hasegawa (1970)	343 to 383
	Boyd et al. (1951)	298
	Fejes and Czárán (1961)	298
	Arai et al. (1967)	313 and 373
	Fuller and Giddings (1967)	302
	Hargrove and Sawyer (1967)	298
	Manner (1967)	298
	Fuller et al. (1969)	302
	Jacobs et al. (1970)	298
N_2 - <i>i</i> -butane	Boyd et al. (1951)	298
N_2 - <i>i</i> -propyl alcohol	Arnikar and Ghule (1969)	358
	Nagata and Hasegawa (1970)	363 and 383
N_2 -nitromethane	Byrne et al. (1967)	303
N_2 - SO_2	Schäfer (1959)	263
N_2 - <i>n</i> -pentane	Arnikar and Ghule (1969)	353
N_2 - <i>n</i> -butylamine	Mehta (1966)	298
N_2 -ethyl formate	Nagata and Hasegawa (1970)	344 to 403
N_2 -methyl acetate	Arnikar et al. (1967 b)	353
	Arnikar and Ghule (1969)	358
	Nagata and Hasegawa (1970)	364 to 403
N_2 -benzene	Bose and Chakraborty (1955-56)	326 and 332
	Hudson et al. (1960)	311
	Arnikar and Ghule (1969)	353
	Nagata and Hasegawa (1970)	364 to 423
N_2 -pyridine	Hudson et al. (1960)	318
N_2 -HBr	Mian (1967)	336 to 525
	Mian et al. (1969)	336 to 525
N_2 -2:3-dimethylbuta-1:3-diene	Cummings et al. (1955)	288
N_2 -thiophen	Hudson et al. (1960)	302
N_2 -cyclohexane	Cummings and Ubbelohde (1953, 1955)	289
	Hudson et al. (1960)	289
	Nagata and Hasegawa (1970)	363 to 403
N_2 -2:3-dimethyl but-2-ene	Cummings et al. (1955)	288
N_2 -methyl cyclopentane	Cummings and Ubbelohde (1953, 1955)	286
N_2 -piperidine	Hudson et al. (1960)	315
N_2 -Rb	McNeal (1962)	328
	Violino (1968)	328
N_2 -3-pentanone	Barr and Sawyer (1964)	300
N_2 -2:3-dimethylbutane	Cummings and Ubbelohde (1953, 1955)	289
N_2 - <i>n</i> -hexane	Cummings and Ubbelohde (1953, 1955)	289
	Huber and van Vught (1965)	353
	Arnikar et al. (1967 b)	353
N_2 -hexa-1:5-diene	Cummings et al. (1955)	288
N_2 -ethyl acetate	Arnikar and Ghule (1969)	355
N_2 -tetrahydrothiophen	Hudson et al. (1960)	319
N_2 - N_2O_4	Chambers and Sherwood (1937)	273 and 283
N_2 -monofluorobenzene	Byrne et al. (1967)	303
N_2 - <i>n</i> -heptane	Cummings et al. (1955)	303
	Clarke and Ubbelohde (1957)	303
N_2 -2:4-dimethylpentane	Clarke and Ubbelohde (1957)	303
N_2 -triethylamine	Mehta (1966)	298
N_2 -Cd	Spier (1940)	290-293

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	T, K
N_2 - <i>n</i> -octane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
N_2 -2:2:4-trimethylpentane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
N_2 - $CHCl_3$	Nagata and Hasegawa (1970)	361 to 418
N_2 - <i>n</i> -nonane	Cummings et al. (1955)	340
N_2 -Cs	Violino (1968)	317
N_2 -1-bromobutane	Byrne et al. (1967)	303
N_2 -2:3:3-trimethylheptane	Cummings and Ubbelohde (1953, 1955)	364
N_2 - <i>n</i> -decane	Cummings and Ubbelohde (1953, 1955)	364
N_2 - SF_6	Suetin and Ivakin (1961)	290
	Ivakin and Suetin (1964 b)	290 to 473
	Suetin (1964)	290
	Evans and Kenney (1965)	293
N_2 -1-bromo-3-methylbutane	Byrne et al. (1967)	303
N_2 - CCl_4	Bose and Chakraborty (1955-56)	323 and 330
	Nagata and Hasegawa (1970)	364 to 423
N_2 - Br_2	Mackenzie and Melville (1932)	286
N_2 -1-iodopropane	Byrne et al. (1967)	303
N_2 - <i>n</i> -dodecane	Cummings and Ubbelohde (1953, 1955)	400
N_2 - Hg	Mullaly and Jacques (1924)	293
	Trautz and Müller (1935)	293
	Spier (1940)	292-298
	Nakayama (1968)	301
N_2 - I_2	Mullaly and Jacques (1924)	293
	Trautz and Müller (1935)	293
	Vyshenskaya and Kosov (1965)	452 to 873
N_2 - UF_6	Ljunggren (1965)	293
CO - CO	Amdur and Shuler (1963)	195 to 373
	Vugts et al. (1970)	233 to 422
CO - C_2H_4	von Obermayer (1883)	290-296
CO - O_2	Weissman (1964)	300 to 550
	Loschmidt (1870 b)	294
	von Obermayer (1883)	287 and 335
	Walker and Westenberg (1960)	295 to 800
	Weissman and Mason (1962 b)	300 to 500
	Walker and Westenberg (1966)	295 to 796
CO - CO_2	Loschmidt (1870 b)	282 and 293
	von Obermayer (1887)	292
	Ivakin and Suetin (1964 a)	296
	Ivakin and Suetin (1964 b)	296 to 473
CO - SF_6	Ivakin and Suetin (1964 a)	297
	Ivakin and Suetin (1964 b)	297 to 473
C_2H_4 - C_2H_4	Mueller and Cahill (1964)	298 to 383
C_2H_4 - C_2H_6	Mueller and Cahill (1964)	298 to 383
C_2H_4 - O_2	Weissman (1964)	293 to 373
C_2H_4 - CO	von Obermayer (1887)	295
C_2H_4 - Rb	McNeal (1962)	333
	Violino (1968)	333
NO - N_2O	Weissman (1964)	550 to 700
C_2H_6 - C_2H_6	Mueller and Cahill (1964)	298 to 383
C_2H_6 - C_3H_8	Weissman (1964)	293 to 523
	Gover (1967)	295
C_2H_6 - CO_2	Gover (1967)	295
C_2H_6 - Rb	McNeal (1962)	333
	Violino (1968)	333
C_2H_6 - <i>n</i> -hexane	Carmichael et al. (1955 b)	294 to 377
O_2 - O_2	Winn (1950)	78 to 353
	Winter (1951)	273 and 318
	Belyaev and Leonas (1966)	2000 to 15 000
O_2 - CO_2	Loschmidt (1870 b)	287
	Wretschko (1870)	297
	von Obermayer (1882 a)	284-293
	Waldmann (1944, 1947)	293
	Walker and Westenberg (1960)	297 to 1080
	Suetin and Ivakin (1961)	288
	Suetin (1964)	288
	Walker and Westenberg (1966)	296 to 1083
	Kosov and Zhalgasov (1970)	202 to 297
O_2 - C_2H_5OH	Bose and Chakraborty (1955-56)	327 and 331
O_2 -benzene	Trautz and Ludwig (1930)	296
	Trautz and Müller (1935)	296
	Bose and Chakraborty (1955-56)	326-332
	Hudson et al. (1960)	311
O_2 -pyridine	Hudson et al. (1960)	318
O_2 -thiophen	Hudson et al. (1960)	302
O_2 -cyclohexane	Cummings and Ubbelohde (1953, 1955)	289
	Hudson et al. (1960)	289
O_2 -methyl cyclopentane	Cummings and Ubbelohde (1953, 1955)	287
O_2 -piperidine	Hudson et al. (1960)	315

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	<i>T</i> , K
O ₂ -2:3-dimethylbutane	Cummings and Ubbelohde (1953, 1955)	288
O ₂ - <i>n</i> -hexane	Cummings and Ubbelohde (1953, 1955)	289
O ₂ -tetrahydrothiophen	Hudson et al. (1960)	319
O ₂ - <i>n</i> -octane	Cummings and Ubbelohde (1953, 1955)	303
O ₂ -2:2:4-trimethylpentane	Cummings and Ubbelohde (1953, 1955)	303
O ₂ -SF ₆	Suetin and Ivakin (1961)	287
	Suetin (1964)	287
	Ivakin et al. (1968)	297 to 408
O ₂ -CCl ₄	Trautz and Müller (1935)	296
	Bose and Chakraborty (1955-56)	323 and 330
O ₂ -Br ₂	Mackenzie and Melville (1932)	286
O ₂ -UF ₆	Ljunggren (1965)	293
air-O ₂	von Obermayer (1882 b)	290-294
	von Obermayer (1887)	287-288
air-CH ₃ OH	Winkelmann (1885)	299 and 323
	Vaillant (1911)	283
	Altshuller and Cohen (1960)	298
	Stevenson (1965)	298
	Getzinger and Wilke (1967)	308
	Lugg (1968)	298
	Mrazek et al. (1968)	328
	Katan (1969)	295
air-H ₂ O ₂	McMurtie and Keyes (1948)	333
air-CO ₂	Loschmidt (1870 a)	252 to 299
	Loschmidt (1870 b)	252 to 291
	von Obermayer (1880)	218-285 and 335
	von Obermayer (1882 a)	284-292
	von Obermayer (1882 b)	283 to 298
	Waitz (1882 a, b)	290-292
	von Obermayer (1883)	280-283 and 335
	von Obermayer (1887)	281-298
	Toepler (1896)	291-292
	Brown and Escombe (1900)	280-288
	Buckingham (1904)	300
	Foch (1913)	Room temperature
	Coward and Georgeson (1937)	273
	Klibanova et al. (1942)	290 to 1533
	Andrew (1955)	291 to 293
	Kosov (1957)	291
	Suetin and Ivakin (1961)	289
	Holsen and Strunk (1964)	276 and 317
	Suetin (1964)	289
air-formic acid	Winkelmann (1885)	339 and 358
	Lugg (1968)	298
air-C ₂ H ₅ OH	Baumgartner (1877 a)	290
	Winkelmann (1884 a)	314 and 340
	Winkelmann (1885)	323 and 337
	Vaillant (1911)	283 to 291
	Le Blanc and Wuppermann (1916)	315 and 340
	Trautz and Müller (1935)	340
	Lee and Wilke (1954)	298
	Bose and Chakraborty (1955-56)	327 and 331
	Narsimhan (1955-56)	303
	Getzinger and Wilke (1967)	308
	Lugg (1968)	298
	Katan (1969)	295
air-acrylonitrile	Lugg (1968)	298
air-acetone	Goryunova and Kuvshinskiĭ (1948)	273
	Gush (1948)	323
	Richardson (1959)	293 to 328
	Stevenson (1965)	298
	Pryde and Pryde (1967)	295
	Lugg (1968)	298
	Lugg (1968)	298
air-allyl alcohol	Winkelmann (1885)	339 to 372
air-acetic acid	Pochettino (1914)	336 to 372
	Lugg (1968)	298
air-methyl formate	Griboiedov (1893)	289-295
	Pochettino (1914)	284 and 293
	Lugg (1968)	298
air- <i>n</i> -propyl alcohol	Winkelmann (1885)	340 and 357
	Vaillant (1911)	287
	Pochettino (1914)	288 to 355
	Lugg (1968)	298
air- <i>i</i> -propyl alcohol	Pochettino (1914)	288 and 333
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
	Lugg (1968)	298
air-ethylene diamine	Klotz and Miller (1947)	273
air-CNCl	Lugg (1968)	298
air-ethylenic glycol	Lugg (1968)	298
air-SO ₂	Andrew (1955)	293

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted*—Continued

d. Other Mixtures—Continued			
System	Reference	T, K	
air-2-methyl-1,3-butadiene air-Cl ₂	Altshuller and Cohen (1960)	288	
	Andrew (1955)	293	
	Kosov (1957)	289	
air-methylethylketone air-pentane	Lugg (1968)	298	
	Lugg (1968)	298	
air-dimethylformamide air- <i>n</i> -butylamine	Lugg (1968)	298	
	Pochettino (1914)	334–335	
air- <i>i</i> -butylamine	Lugg (1968)	298	
	Pochettino (1914)	292 and 335	
air-diethylamine	Lugg (1968)	298	
	Pochettino (1914)	283 and 324	
air-methyl acetate	Lugg (1968)	298	
	Winkelmann (1884 c)	294 and 319	
	Griboiedov (1893)	300 to 317	
	Pochettino (1914)	283 to 324	
air-ethyl formate	Lugg (1968)	298	
	Winkelmann (1884 c)	294 and 319	
	Pochettino (1914)	284 to 323	
air-propionic acid	Lugg (1968)	298	
	Winkelmann (1885)	366 and 372	
	Pochettino (1914)	324 to 372	
air- <i>n</i> -butyl alcohol	Lugg (1968)	298	
	Winkelmann (1885)	372	
	Pochettino (1914)	334 and 373	
	Gilliland (1934)	299 to 332	
air- <i>i</i> -butyl alcohol	Lugg (1968)	298	
	Winkelmann (1885)	340 and 357	
	Pochettino (1914)	333 and 372	
air- <i>sec</i> -butyl alcohol	Lugg (1968)	298	
	Gilliland (1934)	299 to 332	
air- <i>tert</i> -butyl alcohol	Lugg (1968)	298	
	Pochettino (1914)	294 and 340	
air-ethyl ether	Lugg (1968)	298	
	Stefan (1873)	292	
	Baumgartner (1877 a)	289	
	Stefan (1889, 1890)	292	
	Griboiedov (1893)	289–292	
	Winkelmann (1884 a)	284 and 293	
	Naccari (1909, 1910)	285 to 299	
	Pochettino (1914)	283 to 303	
	Trautz and Müller (1935)	288–293	
	Pryde and Pryde (1967)	295	
	Lugg (1968)	298	
	air-CS ₂	Stefan (1873)	289
Baumgartner (1877 a)		291	
Baumgartner (1877 b)		269 to 315	
Stefan (1889, 1890)		289	
Arnold (1944)		303	
Lugg (1968)		298	
air-ethylene glycol-monomethyl ether	Lugg (1968)	298	
	Lugg (1968)	298	
air-propylene glycol	Lugg (1968)	298	
air-allyl chloride	Lugg (1968)	298	
air-benzene	Griboiedov (1893)	315 to 338	
	Le Blanc and Wuppermann (1916)	315 and 340	
	Lee and Wilke (1954)	298	
	Bose and Chakraborty (1955–56)	321 to 332	
	Narsimhan (1955–56)	303	
	Altshuller and Cohen (1960)	300 to 334	
	Jorgensen and Watts (1961)	308	
	Heinzlmann et al. (1965)	308	
	Stevenson (1965)	298	
	Ben Aim et al. (1967)	298	
	Getzinger and Wilke (1967)	308	
	Nafikov and Usmanov (1967)	293 to 333	
	Lugg (1968)	298	
	Grob and El-Wakil (1969)	297	
	Katan (1969)	295	
	Belousova et al. (1970)	300(?)	
	Lugg (1968)	298	
	Goryunova and Kuvshinskiĭ (1948)	318	
	air-cyclohexane	Altshuller and Cohen (1960)	293 and 303
	air-hexene	Lugg (1968)	298
air-dichloromethane	Lugg (1968)	298	
air-methylpropylketone	Lugg (1968)	298	
air- <i>n</i> -hexane	Schlinger et al. (1952–53)	294 to 328	
	Altshuller and Cohen (1960)	298 to 323	
	Galloway and Sage (1967)	311	
	Nafikov and Usmanov (1967)	293 to 333	
	Lugg (1968)	298	
air- <i>n</i> -butyric acid	Grob and El-Wakil (1969)	297	
	Winkelmann (1885)	372	

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	T, K
air- <i>i</i> -butyric acid	Pochettino (1914)	348 and 373
	Lugg (1968)	298
	Winkelmann (1885)	371
air-ethyl acetate	Pochettino (1914)	351 and 373
	Lugg (1968)	298
	Winkelmann (1884 c)	319
air-methyl propionate	Pochettino (1914)	283 to 343
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
	Katan (1969)	295
air-propyl formate	Winkelmann (1884 c)	319 and 340
	Pochettino (1914)	293 and 353
	Lugg (1968)	298
air- <i>p</i> -dioxane	Lugg (1968)	298
air- <i>n</i> -amyl alcohol	Winkelmann (1885)	372
air-active amyl alcohol	Lugg (1968)	298
	Winkelmann (1885)	372
air- <i>sec</i> -amyl alcohol	Gilliland (1934)	299 to 332
air-ethylene glycol- monoethyl ether	Lugg (1968)	298
	Lugg (1968)	298
air-toluene	Mack (1925)	298
air- <i>n</i> -butyl chloride	Gilliland (1934)	299 to 332
	Fairbanks and Wilke (1950)	301
	Narsimhan (1955-56)	303
	Altshuller and Cohen (1960)	298 and 318
	Stevenson (1965)	298
	Yuan and Cheng (1967)	310 to 343
	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
	Mack (1925)	298
	Gilliland (1934)	299 to 332
air-furfural	Lugg (1968)	298
air-fluorobenzene	Brookfield et al. (1947)	298 to 323
air-mesityl oxide	Grob and El-Wakil (1969)	297
air-COCl ₂	Lugg (1968)	298
air-1,1-dichloroethane	Klotz and Miller (1947)	273
air-1,2-dichloroethane	Lugg (1968)	298
	Lugg (1968)	298
air <i>n</i> heptane	Grob and El-Wakil (1969)	297
	Schlinger et al. (1952-53)	294 to 361
	Altshuller and Cohen (1960)	290 and 338
	Stevenson (1965)	298
	Galloway and Sage (1967)	339-350
	Nafikov and Usmanov (1967)	293 to 353
	Grob and El-Wakil (1969)	297
air-triethylamine	Lugg (1968)	298
air- <i>i</i> -butyl formate	Pochettino (1914)	298 to 358
air-ethyl propionate	Lugg (1968)	298
	Winkelmann (1884 c)	340 and 363
	Pochettino (1914)	283 to 366
air-methyl <i>n</i> -butyrate	Fairbanks and Wilke (1950)	301
	Lugg (1968)	298
	Winkelmann (1884 c)	340 and 365
air-methyl <i>i</i> -butyrate	Pochettino (1914)	295 to 372
	Lugg (1968)	298
	Winkelmann (1884 c)	323 and 340
air- <i>n</i> -propyl acetate	Pochettino (1914)	285 and 353
	Lugg (1968)	298
	Le Blanc and Wuppermann (1916)	315 and 340
air- <i>i</i> -propyl acetate	Pochettino (1914)	283 to 372
	Lugg (1968)	298
air-valeric acid	Lugg (1968)	298
air- <i>i</i> -valeric acid	Pochettino (1914)	355 and 373
air-2-ethyl-1-butanol	Winkelmann (1885)	372
	Pochettino (1914)	344 and 373
	Lugg (1968)	298
air- <i>n</i> -hexyl alcohol	Lugg (1968)	298
air- <i>i</i> -propylether	Winkelmann (1885)	372
	Lugg (1968)	298
air-methyl-2-pentanol	Lugg (1968)	298
air-benzonitrile	Lugg (1968)	298
air-phenylethylene	Lugg (1968)	298
air-diethylene glycol	Lugg (1968)	298
air-ethyl benzene	Lugg (1968)	298
	Pochettino (1914)	323 to 373

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted*—Continued

d. Other Mixtures—Continued		
System	Reference	<i>T</i> , K
	Lugg (1968)	298
air- <i>m</i> -xylene	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air- <i>o</i> -xylene	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air- <i>p</i> -xylene	Pochettino (1914)	294 to 373
	Lugg (1968)	298
air-benzyl alcohol	Lugg (1968)	298
air-ethyl bromide	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
air-propylene dichloride	Lugg (1968)	298
air-1-octene	Altshuler and Cohen (1960)	313 and 370
air-chlorobenzene	Le Blanc and Wuppermann (1916)	315 and 340
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air-ethyl cyanoacetate	Lugg (1968)	298
air- <i>n</i> -octane	Mack (1925)	298
	Galloway and Sage (1967)	364
	Nafikov and Usmanov (1967)	293 to 353
	Lugg (1968)	298
air- <i>n</i> -amyl formate	Pochettino (1914)	310 to 373
	Lugg (1968)	298
air- <i>i</i> -amyl formate	Pochettino (1914)	322 and 373
	Lugg (1968)	298
air- <i>n</i> -butyl acetate	Pochettino (1914)	325 and 373
	Lugg (1968)	298
air- <i>i</i> -butyl acetate	Winkelmann (1884 c)	340 and 371
	Pochettino (1914)	324 and 373
	Lugg (1968)	298
air- <i>n</i> -caproic acid	Pochettino (1914)	355 and 373
	Lugg (1968)	298
air- <i>i</i> -caproic acid	Pochettino (1914)	355 and 373
	Lugg (1968)	298
air-diacetone alcohol	Lugg (1968)	298
air-ethyl <i>n</i> -butyrate	Winkelmann (1884 c)	340 and 370
	Pochettino (1914)	315 and 373
	Lugg (1968)	298
air-ethyl <i>i</i> -butyrate	Winkelmann (1884 c)	340 and 369
	Pochettino (1914)	332 and 373
	Lugg (1968)	298
air-methyl valerate	Pochettino (1914)	319 to 373
	Lugg (1968)	298
air-propyl propionate	Winkelmann (1884 c)	370
	Pochettino (1914)	326 to 373
air- <i>n</i> -heptyl alcohol	Lugg (1968)	298
air-CHCl ₃	Baumgartner (1877 a)	292
	Goryunova and Kuyshinskiĭ (1948)	273
	Getzinger and Wilke (1967)	308
	Lugg (1968)	298
	Mrazek et al. (1968)	323
air-mesitylene	Pochettino (1914)	334 and 373
	Lugg (1968)	298
air- <i>n</i> -propyl benzene	Pochettino (1914)	325 to 372
	Lugg (1968)	298
air- <i>i</i> -propyl benzene	Pochettino (1914)	333 to 372
	Lugg (1968)	298
air-pseudo-cumene	Lugg (1968)	298
air-benzoic acid	Yuan and Cheng (1967)	413 to 433
air- <i>n</i> -propyl bromide	Pochettino (1914)	294 and 336
	Lugg (1968)	298
air- <i>i</i> -propyl bromide	Pochettino (1914)	292 and 325
	Lugg (1968)	298
air-nitrobenzene	Lee and Wilke (1954)	298
	Lugg (1968)	298
air-benzyl chloride	Pochettino (1914)	357 and 372
	Lugg (1968)	298
air- <i>o</i> -chlorotoluene	Pochettino (1914)	338 to 371
	Lugg (1968)	298
air- <i>m</i> -chlorotoluene	Pochettino (1914)	338 and 371
	Lugg (1968)	298
air- <i>p</i> -chlorotoluene	Pochettino (1914)	333 to 373
	Lugg (1968)	298
air- <i>n</i> -C ₉ H ₂₀	Nafikov and Usmanov (1967)	293 to 353
air-naphthalene	Mack (1925)	298
air-bromochloromethane	Lugg (1968)	298
air- <i>n</i> -amyl acetate	Lugg (1968)	298
air- <i>n</i> -butyl propionate	Lugg (1968)	298
air- <i>i</i> -butyl propionate	Winkelmann (1884 c)	371
	Pochettino (1914)	329 to 373
	Lugg (1968)	298
air-ethyl valerate	Winkelmann (1884 c)	371

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	T, K
	Pochettino (1914)	324 to 372
air-methyl- <i>n</i> -caproate	Lugg (1968)	298
air- <i>n</i> -propyl <i>n</i> -butyrate	Lugg (1968)	298
	Winkelmann (1884 c)	371
	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air- <i>n</i> -propyl <i>i</i> -butyrate	Winkelmann (1884 c)	370
	Pochettino (1914)	339 to 373
	Lugg (1968)	298
air- <i>i</i> -propyl <i>i</i> -butyrate	Pochettino (1914)	323 and 373
	Lugg (1968)	298
air- <i>n</i> -octyl alcohol	Lugg (1968)	298
air- <i>n</i> -butylether	Lugg (1968)	298
air-trichloro-ethylene	Lugg (1968)	298
air-1,1,1-trichlorethane	Lugg (1968)	298
air-1,1,2-trichlorethane	Lugg (1968)	298
air-diethylene glycol-monoethyl ether	Lugg (1968)	298
air- <i>p</i> -cymene	Lugg (1968)	298
air- <i>i</i> -phorone	Lugg (1968)	298
air-toluene-2,4-diisocyanate	Lugg (1968)	298
air- <i>n</i> -C ₁₀ H ₂₂	Altschuller and Cohen (1960)	313 to 422
	Nafikov and Usmanov (1967)	313 to 353
air-dichloroethylether	Lugg (1968)	298
air- <i>n</i> -amyl propionate	Winkelmann (1884 c)	371
	Pochettino (1914)	323 and 373
	Lugg (1968)	298
air- <i>i</i> -butyl- <i>n</i> -butyrate	Winkelmann (1884 c)	371
	Pochettino (1914)	348 to 373
	Lugg (1968)	298
air- <i>i</i> -butyl- <i>i</i> -butyrate	Winkelmann (1884 c)	371
	Pochettino (1914)	348 and 373
	Lugg (1968)	298
air-propyl valerate	Winkelmann (1884 c)	371
	Pochettino (1914)	343 to 373
	Lugg (1968)	298
air-SF ₆	Suetin and Ivakin (1961)	290
	Suetin (1964)	290
air-ethylene glycol-monoethylether acetate	Lugg (1968)	298
air- <i>n</i> - <i>tert</i> -butyltoluene	Lugg (1968)	298
air-triethylene glycol	Lugg (1968)	298
air-benzyl acetate	Lugg (1968)	298
air-methyl salicylate	Brookfield et al. (1947)	298 and 323
air-CCl ₄	Bose and Chakraborty (1955-56)	323 and 330
	Narsimhan (1955-56)	303
	Richardson (1959)	315 to 335
	Getzinger and Wilke (1967)	308
	Pryde and Pryde (1967)	295
	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
air-diphenyl	Mack (1925)	298
	Gilliland (1934)	491
air- <i>n</i> -C ₁₁ H ₂₄	Nafikov and Usmanov (1967)	333 and 353
air-ethyl iodide	Grob and El-Wakil (1969)	297
air- <i>n</i> -amyl <i>n</i> -butyrate	Pochettino (1914)	324 and 373
	Lugg (1968)	298
air- <i>n</i> -amyl <i>i</i> -butyrate	Winkelmann (1884 c)	371
	Pochettino (1914)	357 and 373
	Lugg (1968)	298
air- <i>i</i> -butyl valerate	Winkelmann (1884 c)	371
	Pochettino (1914)	353 and 373
	Lugg (1968)	298
air-Br ₂	Andrew (1955)	293
	Bröckett (1966)	301
	Lugg (1968)	298
air-safrole	Pochettino (1914)	350 and 373
air- <i>i</i> -safrole	Pochettino (1914)	336 and 373
air-eugenol	Pochettino (1914)	359 and 372
air- <i>i</i> -eugenol	Pochettino (1914)	358 and 372
air-chlorpicrin	Lugg (1968)	298
air-CCl ₃ NO ₂	Klotz and Miller (1947)	298
air-tetrachloroethylene	Lugg (1968)	298
air-1,1,1,2,2-tetrachlorethane	Lugg (1968)	298
air- <i>n</i> -propyl iodide	Pochettino (1914)	304 to 373
	Lugg (1968)	298
air- <i>i</i> -propyl iodide	Pochettino (1914)	324 to 352
	Lugg (1968)	298
air- <i>n</i> -C ₁₂ H ₂₆	Nafikov and Usmanov (1967)	333 and 353
air-anthracene	Mack (1925)	372
air-triethyl phosphate	Lugg (1968)	298

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	T, K
air-benzidine	Mack (1925)	372
air-ethylene dibromide	Call (1957)	273 to 293
	Lugg (1968)	298
air-Hg	Gaede (1915)	Room temperature
	Gilliland (1934)	413 and 473
	Trautz and Müller (1935)	614
	Mikhailov and Kochegarova (1967)	413 and 473
	Lugg (1968)	(?)
	Lugg (1968)	298
air-pentachloroethane	Lugg (1968)	298
air-diethyl phthalate	Lugg (1968)	298
air- <i>n</i> -C ₁₆ H ₃₄	Bradley and Shellard (1949)	288 to 308
air-1,2-dibromo-3-chloropropane	Lugg (1968)	298
air- <i>n</i> -C ₁₇ H ₃₆	Bradley and Shellard (1949)	288 to 313
air-bromoform	Lugg (1968)	298
air-I ₂	Langmuir (1918)	293
	Mack (1925)	298
	Topley and Whytlaw-Gray (1927)	287 to 303
	Trautz and Müller (1935)	287 to 303
	de Nordwall and Flowers (1958)	298
air- <i>n</i> -C ₁₀ H ₂₀	Bradley and Shellard (1949)	288 to 313
air-S ₈	Bradley (1951)	303
air-tributyl phosphate	Lugg (1968)	298
air-di- <i>n</i> -butyl phthalate	Bradley et al. (1946)	293
	Birks and Bradley (1949)	288 to 313
	Lugg (1968)	298
air-tetraethyl pyrophosphate	Lugg (1968)	298
air- <i>bis</i> -2-ethylhexyl phosphate	Lugg (1968)	298
air-(C ₇ H ₁₅) ₃ CH	Bradley and Waghorn (1951)	298 to 308
air-diisooctyl phthalate	Lugg (1968)	298
air-butyl stearate	Bradley et al. (1946)	293
air- <i>tri</i> -orthocresol phosphate	Lugg (1968)	298
air-(C ₁₀ H ₂₁) ₃ CH	Bradley and Waghorn (1951)	298 to 308
air- <i>n</i> -C ₁₆ F ₃₄	Bradley and Waghorn (1951)	288 to 303
CH ₃ OH-CO ₂	Winkelmann (1885)	299 and 323
CH ₃ OH-1-butanol	Weissmann (1968 a)	423
H ₂ S-ethyl ether	Pal and Bhattacharyya (1969)	298 to 373
HCl-DCl	Braune and Zehle (1941)	293–296
HCl-CO ₂	Weissman (1964)	291
HCl-Br ₂	Mackenzie and Melville (1933)	288
C ₃ H ₆ -C ₃ H ₈	Weissmann (1964)	313 to 373
CO ₂ -CO ₂	Winn (1950)	195 to 353
	Timmerhaus and Drickamer (1951)	296–297
	Winter (1951)	273 and 318
	Amdur et al. (1952)	195 to 363
	Miller and Carman (1961)	293
	Ember et al. (1962)	297, 1180 to 1680
	Schäfer and Reinhard (1963)	233 to 513
	Wendt et al. (1963)	248 to 362
	Ember et al. (1964)	297
	Miller and Carman (1964)	293
	Pakurar and Ferron (1964)	295, 1250 to 1650
	Pakurar and Ferron (1965)	1103 to 1944
	Ferron (1967)	300 to 1900
	Annis et al. (1969)	295
	Mistler et al. (1970)	293
CO ₂ -N ₂ O	Loschmidt (1870 b)	288 and 293
	von Obermayer (1880)	283–285 and 335
	von Obermayer (1882 b)	283–287
	Boardman and Wild (1937)	286–287
	Wall and Kidder (1946)	298
	Amdur et al. (1952)	195 to 363
	Weissman (1964)	300 to 550
	Kosov and Abdullina (1966)	298
CO ₂ -ethylene oxide	Wall and Kidder (1946)	298
CO ₂ -C ₃ H ₈	Wall and Kidder (1946)	298
	Weissman (1964)	300 to 550
CO ₂ -formic acid	Winkelmann (1885)	339 and 358
CO ₂ -C ₂ H ₅ OH	Winkelmann (1884 a)	314 and 340
	Winkelmann (1885)	323 and 337
	Trautz and Müller (1935)	315 and 340
CO ₂ -acetic acid	Winkelmann (1885)	339 to 372
CO ₂ - <i>n</i> -propyl alcohol	Winkelmann (1885)	340 and 357
CO ₂ - <i>i</i> -propyl alcohol	Nagata and Hasegawa (1970)	363 to 418
CO ₂ -SO ₂	Schäfer (1959)	263 to 473
	Weissman (1964)	289
CO ₂ -ethyl formate	Winkelmann (1884 c)	294 and 319
	Nagata and Hasegawa (1970)	334 to 363
CO ₂ -methyl acetate	Winkelmann (1884 c)	294 and 319
	Nagata and Hasegawa (1970)	363 and 383
CO ₂ -propionic acid	Winkelmann (1885)	366 and 372

TABLE 16. Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued

d. Other Mixtures—Continued		
System	Reference	T, K
CO ₂ - <i>n</i> -butyl alcohol	Winkelmann (1885)	372
CO ₂ - <i>i</i> -butyl alcohol	Winkelmann (1885)	340 and 357
CO ₂ -ethyl ether	Baumgartner (1877 a)	291
	Winkelmann (1884 a)	283 and 293
	Trautz and Müller (1935)	290–293
CO ₂ -CS ₂	Baumgartner (1877 a)	290
	Baumgartner (1877 b)	267 to 313
CO ₂ -benzene	Nagata and Hasegawa (1970)	364 to 423
CO ₂ -cyclohexane	Nagata and Hasegawa (1970)	363 to 423
CO ₂ - <i>n</i> -butyric acid	Winkelmann (1885)	372
CO ₂ - <i>i</i> -butyric acid	Winkelmann (1885)	371
CO ₂ -ethyl acetate	Winkelmann (1884 c)	319
CO ₂ -methyl propionate	Winkelmann (1884 c)	319 and 340
CO ₂ -propyl formate	Winkelmann (1884 c)	319 and 340
CO ₂ - <i>n</i> -amyl alcohol	Winkelmann (1885)	372
CO ₂ -active amyl alcohol	Winkelmann (1885)	372
CO ₂ -ethyl propionate	Winkelmann (1884 c)	340 and 363
CO ₂ -methyl butyrate	Winkelmann (1884 c)	340 and 365
CO ₂ -methyl <i>i</i> -butyrate	Winkelmann (1884 c)	323 and 340
CO ₂ - <i>i</i> -valeric acid	Winkelmann (1885)	372
CO ₂ - <i>n</i> -hexyl alcohol	Winkelmann (1885)	372
CO ₂ - <i>i</i> -butyl acetate	Winkelmann (1884 c)	340 and 371
CO ₂ -ethyl butyrate	Winkelmann (1884 c)	340 and 370
CO ₂ -ethyl <i>i</i> -butyrate	Winkelmann (1884 c)	340 and 369
CO ₂ -ethyl valerate	Winkelmann (1884 c)	371
CO ₂ -propyl propionate	Winkelmann (1884 c)	370
CO ₂ -CHCl ₃	Baumgartner (1877 a)	291
	Nagata and Hasegawa (1970)	363 to 404
CO ₂ - <i>i</i> -butyl propionate	Winkelmann (1884 c)	371
CO ₂ -propyl butyrate	Winkelmann (1884 c)	371
CO ₂ -propyl- <i>i</i> -butyrate	Winkelmann (1884 c)	370
CO ₂ -amyl propionate	Winkelmann (1884 c)	371
CO ₂ - <i>i</i> -butyl butyrate	Winkelmann (1884 c)	371
CO ₂ - <i>i</i> -butyl- <i>i</i> -butyrate	Winkelmann (1884 c)	371
CO ₂ -propyl valerate	Winkelmann (1884 c)	371
CO ₂ -SF ₆	Suetin and Ivakin (1961)	291
	Suetin (1964)	291
	Ivakin and Suetin (1964 b)	291 to 472
CO ₂ -CCl ₄	Nagata and Hasegawa (1970)	363 to 423
CO ₂ -amyl <i>i</i> -butyrate	Winkelmann (1884 c)	371
CO ₂ - <i>i</i> -butyl valerate	Winkelmann (1884 c)	371
CO ₂ -Br ₂	Mackenzie and Melville (1932)	288
	Mackenzie and Melville (1933)	290
CO ₂ -I ₂	Vyshenskaya and Kosov (1965)	452 to 1275
N ₂ O-ethylene oxide	Wall and Kidder (1946)	298
N ₂ O-C ₃ H ₈	Wall and Kidder (1946)	298
	Weissman (1964)	300 to 550
N ₂ O-Br ₂	Mackenzie and Melville (1933)	290
C ₆ H ₆ - <i>n</i> -hexane	Carmichael et al. (1955 b)	294 to 377
NO ₂ -N ₂ O ₄	Weissman (1968 a)	303 to 343
C ₂ H ₅ OH-C ₄ H ₉ OH	Weissman (1968 a)	423
C ₂ H ₅ OH-CCl ₂ F ₂	Lee and Wilke (1954)	298
(CH ₃) ₂ O-CH ₃ Cl	Chakraborti and Gray (1966)	303 to 333
	Weissman (1968 a)	308 and 353
(CH ₃) ₂ O-SO ₂	Chakraborti and Gray (1966)	303 to 333
	Weissman (1968 a)	308 and 353
CH ₃ Cl-SO ₂	Chakraborti and Gray (1966)	303 to 333
	Weissman (1968 a)	308 and 353
CH ₃ Cl-C ₂ H ₅ Cl	Manner (1967)	298 to 419
1,3-butadiene-1-butyne	Bournia et al. (1961)	300
C ₃ H ₇ OH-C ₄ H ₉ OH	Weissman (1968 a)	423
SO ₂ -SO ₂ F ₂	Chang et al. (1970)	273 to 673
BF ₃ -BF ₃	Zmbov and Knežević (1961)	298 and 316
BF ₃ -CCl ₄	Raw (1955)	303
<i>n</i> -C ₅ H ₁₂ - <i>n</i> -C ₅ H ₁₂	Beatty (1969)	273
<i>n</i> -C ₅ H ₁₂ -C(CH ₃) ₄	Beatty (1969)	273
(C ₂ H ₅) ₂ O-CHCl ₃	Weissman (1964)	293
C ₆ H ₆ -CCl ₄	Weissman (1964)	293
C ₆ H ₆ -CCl ₂ F ₂	Lee and Wilke (1954)	298
HBr-DBr	Braune and Zehle (1941)	294–296
cyclohexane-Rb	McNeal (1962)	323
	Violino (1968)	323
	Weissman (1964)	293 to 413
CH ₂ Cl ₂ -CCl ₄	Khoury and Kobayashi (1970)	243 to 348
CF ₄ -CF ₄	Raw and Tang (1963)	303 to 342
CF ₄ -SF ₆	Raw and Tang (1963)	303 to 342
<i>i</i> -octane-C ₆ H ₅ NO ₂	Huber and van Vught (1965)	298
triethylamine-CCl ₂ F ₂	Mehta (1966)	298
C ₈ H ₁₈ -C ₇ F ₁₆	Weissman (1964)	303 and 323
BCl ₃ -CCl ₄	Raw (1955)	303
CHCl ₃ -CCl ₄	Weissman (1964)	293
Cl ₂ CF ₂ -Cl ₂ CF ₂	Miller and Carman (1961)	293

TABLE 16. *Experimental determinations of \mathcal{D}_{12} according to gas pair, temperature noted—Continued*

d. Other Mixtures—Continued		
System	Reference	<i>T</i> , K
CCl ₂ F ₂ -di- <i>n</i> -butyl phthalate Hg-Hg	Birks and Bradley (1949)	293 and 303
	Coulliette (1928)	338 to 376
	Biondi (1953)	350
	McCoubrey (1954)	473
	McCoubrey and Matland (1954)	473(?)
	Matland and McCoubrey (1955)	380 to 580
	McCoubrey and Matland (1956)	473
Hg-I ₂	Mullaly and Jacques (1924)	292
GeBr ₂ -GeBr ₄	Jona (1965)	684
GeI ₂ -GeI ₄	Jona (1965)	684
UF ₆ -UF ₆	Ney and Armistead (1947)	297–301
	Brown and Murphy (1965)	273 to 344

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6. Acknowledgments

This work is based on the Ph.D. dissertation of T. R. Marrero at the University of Maryland. It was supported in part by National Aeronautics and Space Administration Grant NGR-21-002-053 to the University of Maryland, and in part by Army Research Office (Durham) Grant DA-ARO-D-31-124-G925 to Brown University. The first author thanks Truman S. Storvick, University of Missouri—Columbia, for his support during the proofreading phase of this work.

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*A misprint appears in table 1; the highest temperature for CH₄-H₂O is not 323 K but 328 K.

- O₂, air, CO₂, SF₆), Ar-(C₂H₂, CO₂, SF₆), H₂-(He, Ar, N₂, air, CO₂, SF₆), N₂-(CO₂, SF₆), O₂-(C₂H₂, CO₂, SF₆), air-(CO₂, SF₆), N₂-(CO₂, SF₆), O₂-(acetylene, CO₂, SF₆), air-(CO₂, acetylene, SF₆), CO₂-SF₆, closed tube.
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- Winkelmann, A. (1884 c), *Ann. Physik* **23**, 203; H₂, air, and CO₂ (each with the esters as follows): (ethyl formate, propyl formate, methyl acetate, ethyl acetate, *i*-butyl acetate, methyl propionate, ethyl propionate, propyl propionate, *i*-butyl propionate, amyl propionate, methyl butyrate, ethyl butyrate, propyl butyrate, *i*-butyl butyrate, methyl-*i*-butyrate, ethyl-*i*-butyrate, propyl-*i*-butyrate, *i*-butyl-*i*-butyrate, amyl-*i*-butyrate, ethyl valerate, propyl valerate, *i*-butyl valerate), evaporation tube.
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The gas pairs investigated and the potential energy separation ranges are noted.

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2. Moscow State University (Leonas et al.)

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4. Reviews

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Bibliography III. London Dispersion Constants

Dalgarno, A. (1967), *Adv. Chem. Phys.* **12**, 143. This is a comprehensive review and summary.

Bibliography IV. Related Viscosity Measurements

The gas pairs investigated and the temperature ranges are noted.

1. Primary References

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2. Secondary References

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- Weissman, S. (1964), *J. Chem. Phys.* **40**, 3397. See Bibliography I.
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