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Gaseous Diffusion Coefficients

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Gaseous Diffusion Coefficients

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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 cm^3). 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

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[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.

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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 understanting 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 multicomponent 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

$$\mathbf{J}_1 = -n\mathscr{D}_{12}\nabla x_1, \qquad (2.1\text{–}1)$$

$$\mathbf{J}_2 = -n\mathscr{D}_{21}\nabla x_2. \qquad (2.1\text{–}2)$$

Each species (or component) is labeled by subscripts 1 or 2. The flux densities are $\mathbf{J}_1$ and $\mathbf{J}_2$ (molecules/cm$^2\cdot$s), the total number density is n (molecules/cm^3), 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 $\mathscr{D}_{12}$ and $\mathscr{D}_{21}$ are positive constants with units of cm^2/s. It is easy to show from eqs (2.1–1) and (2.1–2) that $\mathscr{D}_{12}=\mathscr{D}_{21}$, 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

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[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

$$\mathbf{J}_1 = -n\mathscr{D}_{12}\nabla x_1 + x_1\mathbf{J}, \qquad (2.1\text{–}3)$$

$$\mathbf{J}_2 = -n\mathscr{D}_{21}\nabla x_2 + x_2\mathbf{J}, \qquad (2.1\text{–}4)$$

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 $\mathscr{D}_{12} = \mathscr{D}_{21}$.

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

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

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 $\mathscr{D}'_{ij}$ depend primarily only on the nature of the species i and j, but are not quite the same as the corresponding binary diffusion coefficients $\mathscr{D}_{ij}$ (hence the prime). However, the difference between the $\mathscr{D}'_{ij}$ and the $\mathscr{D}_{ij}$ lies only in their weak composition dependence, the exact value of $\mathscr{D}'_{ij}$ 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 $\mathscr{D}_{ij}$ with composition is empirically a few percent at most, and is of the same magnitude as the experimental uncertainties in the few available multicomponent $\mathscr{D}'_{ij}$, or even in most measured binary $\mathscr{D}_{ij}$ for that matter. Therefore it is reasonable on an empirical basis to take $\mathscr{D}'_{ij} \approx \mathscr{D}_{ij}$ for multicomponent diffusion. This is also justified theoretically, for in the first Chapman-Enskog approximation $\mathscr{D}'_{ij}$ and $\mathscr{D}_{ij}$ 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 $\mathscr{D}'_{ij} \approx \mathscr{D}_{ij}$, 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,

$$\nabla x_1 = -\mathbf{J}_1 \sum_{j=2}^{\nu} \frac{x_j}{n\mathscr{D}'_{ij}}. \qquad (2.1\text{–}6)$$

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

$$\frac{1}{\mathscr{D}_1} = \sum_{j=2}^{\nu} \frac{x_j}{\mathscr{D}'_{ij}}, \qquad (2.1\text{–}7)$$

where $\mathscr{D}_1$ specifies the diffusion coefficient of the tracer in the multicomponent mixture. If $\mathscr{D}_{ij}$ replaces $\mathscr{D}'_{ij}$ then eq (2.1–7) becomes an expression of Blanc's law [28]. Detailed calculations [29] of $\mathscr{D}'_{ij}$ and $\mathscr{D}_{ij}$ 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 integro-differential 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 integro-differential 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

$$[\mathscr{D}_{12}]_M=[\mathscr{D}_{12}]_1 f^{(M)}, \qquad (2.2\text{–}1)$$

where $[\mathscr{D}_{12}]_1$ 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

$$f^{(M)}=1/(1-\Delta_{12}-\ldots), \qquad (2.2\text{–}2a)$$

or

$$f^{(M)}=(1+\Delta_{12}+\ldots), \qquad (2.2\text{–}2b)$$

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

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

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

where $\mu_{12}=m_1m_2/(m_1+m_2)$ 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

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

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

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

where $I(\chi, E)$ is the differential scattering cross section. For classical systems $I(\chi, E)\sin\chi d\chi=bdb$, 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

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

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

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

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

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

$$[\mathscr{D}_{12}]_1=0.008258\left(\frac{M_1+M_2}{2M_1M_2}\right)^{1/2}\frac{T^{3/2}}{p\bar{\Omega}_{12}^{(1,1)}}, \qquad (2.2\text{–}8)$$

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

$$[\mathscr{D}_{12}]_2\equiv[\mathscr{D}_{12}]_1(1+\Delta_{12}), \qquad (2.2\text{–}9)$$

where

$$\Delta_{12}-\frac{(6C_{12}^*-5)^2}{10}\left(\frac{x_1^2P_1+x_2^2P_2+x_1x_2P_{12}}{x_1^2Q_1+x_2^2Q_2+x_1x_2Q_{12}}\right). \qquad (2.2\text{–}10)$$

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 $[\mathscr{D}_{12}]_1$ is to $\lim_{M\to\infty}[\mathscr{D}_{12}]_M$ depends on composition, molecular masses, and the intermolecular forces of the gas mixture. Of course, an experiment measures only $\lim_{M\to\infty}[\mathscr{D}_{12}]_M$. By numerical comparison of $[\mathscr{D}_{12}]_1$, $[\mathscr{D}_{12}]_2$, $[\mathscr{D}_{12}]_3$, etc. for a variety of special cases, the accuracy of $[\mathscr{D}_{12}]_1$ may be assessed [30]. For the case of nearly equal molecular masses $[\mathscr{D}_{12}]_1$ 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 $[\mathscr{D}_{12}]_1$ is accurate to within 1 percent.

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

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

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

with

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

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

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

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

$$A^* = \Omega^{(2,2)*}/\Omega^{(1,1)*}, \qquad (2.2\text{–}14)$$

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

$$C^* = \Omega^{(1,2)*}/\Omega^{(1,1)*}, \qquad (2.2\text{–}16)$$

$$E^* = \Omega^{(2,3)*}/\Omega^{(2,2)*}. \qquad (2.2\text{–}17)$$

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

$$T^* \equiv kT/\epsilon, \qquad (2.2\text{–}18)$$

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:

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

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

where

$$\gamma^2 S_{ij}^{(1)kl}(E)=\int_0^{2\pi} d\phi \int_0^\pi I_{ij}^{kl}(\chi,\phi,E)$$

$$\sin\chi d\chi(\gamma^2-\gamma\gamma'\cos\chi), \qquad (2.2\text{–}20)$$

$$\gamma^4 S_{ij}^{(2)kl}(E)=\tfrac{3}{2}\int_0^{2\pi} d\phi \int_0^\pi I_{ij}^{kl}(\chi,\phi,E)$$

$$\sin\chi d\chi[\gamma^2(\gamma^2-\gamma'^2\cos^2\chi)-\tfrac{1}{6}(\gamma^2-\gamma'^2)^2], \qquad (2.2\text{–}21)$$

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

$$\gamma^2=E/kT, \qquad (2.2\text{–}23a)$$

$$\gamma'^2=E'/kT, \qquad (2.2\text{–}23b)$$

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: $Z_q=\Sigma \exp(-\epsilon_{qi})$ and $Z_{q'}=\Sigma\exp(-\epsilon_{q'j})$. They appear only as normalization factors in eq (2.2–19). The indices i and j denote the ith and jth internal quantum states of the qth 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 lth 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $[\mathscr{D}_{12}]_1$; 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 $[\mathscr{D}_{12}]_1$ 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 $(\partial\ln\mathscr{D}_{12}/\partial\ln T)_p$ lies between 3/2 and 2. Thus $\mathscr{D}_{12}$ 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.

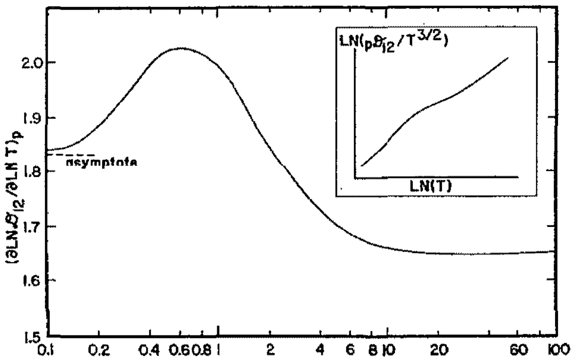

FIGURE 1. *Qualitative temperature dependence of diffusion coefficients.*

This figure shows the derivative $(\partial\ln\mathscr{D}_{12}/\partial\ln T)_p$, 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 $(\partial\ln\mathscr{D}_{12}/\partial\ln T)_p$ 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 $(\partial\ln\mathscr{D}_{12}/\partial\ln T)_p$ 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 $(\partial\ln\mathscr{D}_{12}/\partial\ln T)_p$ is not monotonic, and exhibits a maximum where both short-range and long-range forces are significant.

In figure 1 the inset shows $\ln(p\mathscr{D}_{12}/T^{3/2})$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ *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 $\mathscr{D}_{12}$ are considered below.

a. Long-Range Interactions

These interactions behave asymptotically as (neglecting retardation effects)

$$\varphi(r) = -C/r^6, \tag{2.3–1}$$

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

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

Thus as $T \to 0$, $\mathscr{D}_{12} \propto T^{11/6}$ classically, but at sufficiently low temperatures quantum corrections *become important. A general expression for the* quantum-mechanical $\bar{\Omega}^{(1,1)}$ as $T \to 0$ 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 $\mathscr{D}_{12}$ at high temperatures, $T \gtrsim 1000$ K.

The expression for the exponential potential is

$$\varphi(r) = \varphi_0 \exp(-r/\rho), \tag{2.3–3}$$

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

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

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

$$\varphi(r) = K/r^s, \tag{2.3–5}$$

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

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

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 $(n-6)$

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

and

Exp–6

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

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

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

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

$$\mathscr{D}_{12} \propto T^{3/2}/(1+S/T), \tag{2.3–10}$$

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

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

or

$$\mathscr{D}_{12} \propto T^{3/2} e^{-S/T}, \tag{2.3–12}$$

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 $\mathscr{D}_{12}$ measurements. The composition correction term, Δ_{12}, is repeated here for convenience, and the P and Q terms are expressed as follows:

$$\Delta_{12}=\frac{(6C_{12}^{*}-5)^2}{10}\left(\frac{x_1^2P_1+x_2^2P_2+x_1x_2P_{12}}{x_1^2Q_1+x_2^2Q_2+x_1x_2Q_{12}}\right), \tag{2.2–10}$$

where

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

$$P_{12}=15\left(\frac{M_1-M_2}{M_1+M_2}\right)^2+\frac{8M_1M_2A_{12}^{*}}{(M_1+M_2)^2}, \tag{2.4–2}$$

$$Q_1=\frac{2}{M_2(M_1+M_2)}\left(\frac{2M_2}{M_1+M_2}\right)^{1/2}\frac{\Omega_{11}^{(2,2)*}}{\Omega_{12}^{(1,1)*}}\left(\frac{\sigma_{11}}{\sigma_{12}}\right)^2 \times\left[\left(\frac{5}{2}-\frac{6}{5}B_{12}^{*}\right)M_1^2+3M_2^2+\frac{8}{5}M_1M_2A_{12}^{*}\right], \tag{2.4–3}$$

$$Q_{12}=15\left(\frac{M_1-M_2}{M_1+M_2}\right)^2\left(\frac{5}{2}-\frac{6}{5}B_{12}^{*}\right)+\frac{4M_1M_2A_{12}^{*}}{(M_1+M_2)^2}\left(11-\frac{12}{5}B_{12}^{*}\right)+\frac{8}{5}\frac{(M_1+M_2)}{(M_1M_2)^{1/2}}\frac{\Omega_{11}^{(2,2)*}}{\Omega_{12}^{(1,1)*}}\frac{\Omega_{22}^{(2,2)*}}{\Omega_{12}^{(1,1)*}}\left(\frac{\sigma_{11}}{\sigma_{12}}\right)^2\left(\frac{\sigma_{22}}{\sigma_{12}}\right)^2. \tag{2.4–4}$$

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^*,

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

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 H_2-D_2, and 2.9 for ^{3}He-^{4}He. From table 1 it is evident that quantum deviations in $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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

$$I(\chi)=|f(\chi)|^2, \tag{2.5–2}$$

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

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]

	$[\mathscr{D}_{12}]_1$ (Quantal)/$[\mathscr{D}_{12}]_1$ (Classical)						A_{12}^{*} (Quantal)/A_{12}^{*} (Classical)				
T^* \ Λ^*	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:

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

and

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

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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 quantal 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 Bibilography, sections II and III.

a. Diffusion Collision Integrals for Long-Range Interactions

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

$$\bar{\Omega}^{(1,1)}=71.1(C/T)^{1/3}, \qquad (2.6\text{–}1)$$

in which C is the London constant in atomic units $(e^2a_0^5)$ 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, $[\mathscr{D}_{12}]_1$ 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 $[\mathscr{D}_{12}]_1$ 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 $\lesssim 0.2$. 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 $T^* \geqslant 0.1$, eq (2.6–1) is useful only to a 10 percent level of uncertainty. For $\Lambda^*=0.5$ and $T^* \geqslant 0.02$ 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

$$\bar{\Omega}^{(1,1)}=4\pi\alpha^2\rho^2 I_{(1,1)}, \qquad (2.6\text{–}2)$$

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

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

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 $\mathscr{D}_{12}$ 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 He-Ar, He-N_2, and H_2-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 $\mathscr{D}_{12}$ 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,

$$C_{12}=(C_{11}C_{22})^{1/2}. \qquad (2.6\text{–}4)$$

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

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

$$\rho_{12}^{-1}=\tfrac{1}{2}(\rho_{11}^{-1}+\rho_{22}^{-1}), \qquad (2.6\text{–}5b)$$

and

Inverse-Power Potential

$$K_{12}=(K_{11}K_{22})^{1/2}, \qquad (2.6\text{–}6a)$$

$$s_{12}=\tfrac{1}{2}(s_{11}+s_{22}). \qquad (2.6\text{–}6b)$$

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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$.

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:

$$(p\mathscr{D}_{12})^2a+(p\mathscr{D}_{12})b + (p\mathscr{D}_{12})cA_{12}^*+A_{12}^*d=0, \qquad (2.7\text{–}1)$$

in which

$$a=(x_1x_2)^2(\eta_{\text{mix}}-\eta_1-\eta_2)/\eta_1\eta_2, \qquad (2.7\text{–}2)$$

$$b=2x_1x_2(M_1+M_2)^{-1}RT[\eta_{\text{mix}}(x_1^2\eta_2+x_1^2\eta_1) -\eta_1\eta_2]/\eta_1\eta_2, \qquad (2.7\text{–}3)$$

$$c=\frac{6}{5}x_1x_2(M_1+M_2)^{-1}RT[\eta_{\text{mix}}(x_1^2M_1^2\eta_2+x_2^2M_2^2\eta_1) -(x_1M_1-x_2M_2)^2\eta_1\eta_2]/M_1M_2\eta_1\eta_2, \qquad (2.7\text{–}4)$$

$$d=\frac{3}{5}(2x_1x_2RT)^2\eta_{\mathrm{mix}}/M_1M_2, \qquad (2.7\text{–}5)$$

where R is the gas constant (82.0567 $\mathrm{cm}^3\cdot\mathrm{atm}/\mathrm{mole}\cdot\mathrm{K}$), η is the viscosity in g/cm · s, η_{mix} denotes the mixture viscosity, and the subscripts have their usual meaning. The determination of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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

$$p\mathscr{D}_{11}=\frac{6}{5}A_{11}^*(RT/M_1)\eta_1, \qquad (2.7\text{–}6)$$

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

The determination of $\mathscr{D}_{12}$ from viscosity measurements has been derived from *first approximation* formulas. On this basis the diffusion coefficients calculated cannot be the true values of $\mathscr{D}_{12}$, which have a small composition dependence. The diffusion coefficients calculated cannot be exact $[\mathscr{D}_{12}]_1$ because experimental viscosity data are used [58]. However, the diffusion coefficients calculated from experimental binary mixture viscosity data are nearly equal to $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ calculated from mixture viscosity measurements is almost the same as obtainable by $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ measurements at the corresponding temperatures. Second, remove the restriction of a perfectly known A_{12}^* in order to obtain the total uncertainty of calculated $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$, that is, a 1 percent error in A_{12}^* corresponds to an error of approximately 1 percent in $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ [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 $\mathscr{D}_{12}$ measurements for two reasons. First, the relationship between thermal conductivity and $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ at the same temperature (the value of A_{12}^* is still assigned theoretically). Since B_{12}^* is related to the temperature derivative of $\mathscr{D}_{12}$, the temperature range of $\mathscr{D}_{12}$ can be extended. That is, if λ_{mix} and $\mathscr{D}_{12}$ are known at a single temperature, values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$ at present. Another procedure relates the temperature dependence of $\mathscr{D}_{12}$ to that of α_T, and the derived relationship is combined with a single measurement of $\mathscr{D}_{12}$ to produce diffusion coefficients over a wide temperature range [61]. From this procedure the accuracy of $\mathscr{D}_{12}$ 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

$$(6C_{12}^*-5)=2[2-(\partial \ln [\mathscr{D}_{12}]_1/\partial \ln T)_p], \qquad (2.7\text{–}7)$$

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

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

$$-S_2=\frac{15}{2}\frac{M_1(M_1-M_2)}{(M_1+M_2)^2}+4\frac{M_1M_2}{(M_1+M_2)^2}A_{12}^{*}$$

$$-\frac{5}{3}\frac{M_2^2}{(M_1+M_2)}\frac{p[\mathscr{D}_{12}]_1}{[\eta_2]_1RT}, \tag{2.7–9}$$

$$Q_2=\frac{10}{3}\frac{M_2}{(M_1+M_2)^2}\frac{p[\mathscr{D}_{12}]_1}{[\eta_2]_1RT}(3M_1^2$$

$$+M_2^2+\frac{8}{5}M_1M_2A_{12}^{*}), \tag{2.7–10}$$

$$\kappa_2=\frac{1}{42}(8E_{22}^{*}-7)^2+\frac{2}{21}\left(1-\frac{M_2}{M_1}\right)(8E_{22}^{*}-7)$$

$$\left[1-\frac{3}{4}(5-4B_{12}^{*})(6C_{12}^{*}-5)^{-1}\right], \tag{2.7–11}$$

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

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 $[\mathscr{D}_{12}]_1$ 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 interation 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 $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$. The result is a relationship for the temperature dependence of $\mathscr{D}_{12}$ over the range for which measurements of α_T are available. The second iteration step uses values of $[\mathscr{D}_{12}]_1$ 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 $[\mathscr{D}_{12}]_1$ by the integration of eq (2.7–7). The $[\mathscr{D}_{12}]_1$ 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 $\mathscr{D}_{12}$; 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 diffusion 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 $\mathscr{D}_{12}$ 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	Hartcck 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); Krongelb 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.

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

oped; their results include almost all values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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.

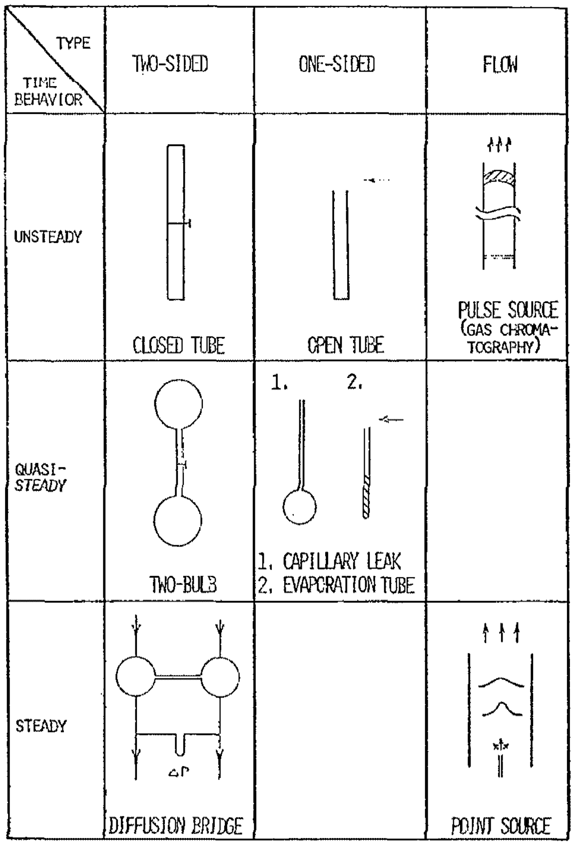

FIGURE 2. *Principal experimental methods for diffusion coefficients.*

3.2 Major Experimental Methods

a. Closed Tube

In 1870 the closed-tube method was developed by Loschmidt, who carefully determined $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 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,

$$\partial x_1/\partial t=\mathscr{D}_{12}(\partial^2 x_1/\partial z^2), \qquad (3.2\text{–}1)$$

where $\mathscr{D}_{12}$ 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,

$$x_1=x_1^l \qquad 0\leqslant z<L/2,\ t=0$$

$$x_1=x_1^u \qquad L/2<z<L,\ t=0$$

$$\partial x_1/\partial z=0 \qquad z=0 \text{ and } z=L,\ t\geqslant 0,$$

the solution of eq (3.2–1) is

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

and the relaxation time is

$$\tau=L^2/\pi^2\mathscr{D}_{12}, \qquad (3.2\text{–}3)$$

TABLE 3. *Determinations of $\mathscr{D}_{12}$ 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
Groth and Harteck	1941	Ljunggren	1965
Heath et al	1941	Reichenbacher et al	1965
Groth 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ on the mixture composition. The diffusion coefficient was assumed independent of mixture composition. Equation (3.2–1) rewritten to express the composition dependence of $\mathscr{D}_{12}$ is

$$\partial x_1/\partial t = \mathscr{D}_{12}(\partial^2 x_1/\partial z^2) + (\partial x_1/\partial z)^2(\partial \mathscr{D}_{12}/\partial x_1). \qquad (3.2\text{–}4)$$

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 $\mathscr{D}_{12}$ may be significant. However, for the case of the closed tube, the exact integration of the diffusion equation with a composition-dependent $\mathscr{D}_{12}$ has not been performed. The uncertainty of results caused by the composition dependence of $\mathscr{D}_{12}$ depends on the duration of the experimental run, the gas pair investigated, and the initial composition of the mixture. Calculated values of $\mathscr{D}_{12}$ would apparently depend on the length of the experimental run. This has been investigated [15, 18], and the variation of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ often have reproducibilities better than 1 percent, and the measurements have been reported accurate to 1 to 3 percent. However, independent determinations of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ can be found from the relaxation time.

The determinations of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$, 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 $\mathscr{D}_{12}$ 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

$$\Delta x(t) = \Delta x(0) \exp(-t/\tau), \qquad (3.2\text{–}5)$$

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

$$\tau = \frac{1}{\mathscr{D}_{12}} \left(\frac{L}{A}\right) \left(\frac{V_1 V_2}{V_1 + V_2}\right), \qquad (3.2\text{–}6)$$

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 $\mathscr{D}_{12}$ 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,

$$\tau = \frac{K}{\mathscr{D}_{12}}\left(\frac{L}{A}\right)\left(\frac{V_1V_2}{V_1+V_2}\right), \qquad (3.2\text{–}7)$$

where

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

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

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

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 $\alpha \gtrsim 0.82$ 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 $\Sigma_j(L_j/A_j)$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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

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

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:

$$\lim_{R\to\infty} x = 0,$$

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

and the normalization condition

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

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 $\mathscr{D}_{12}$. 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

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

A concentration profile can be used to determine $\mathscr{D}_{12}$, 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 $-U/2\mathscr{D}_{12}$, and measurements of the carrier velocity U lead to values of $\mathscr{D}_{12}$. Alternately, the stream can be sampled at points on the axis, $z = R$, and values of $\mathscr{D}_{12}$ calculated from

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

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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$.

The advantages of the gas-chromatography method are as follows. Determinations of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ is practical because of the small amount of sample required to make a pulse. These advantages are operational, only.

Determinations of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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
Zhukhovitskiĭ 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 $\mathscr{D}_{12}$.

Packed chromatography columns have been used, on occasion, to determine $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$, the theory had been developed for diffusion phenomena in the flow of fluids [29–34]. The basic equation for the gas-chromatography method is

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

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 $\mathscr{D}_{12}$, 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

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

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 $\mathscr{D}_{\mathrm{eff}}$ denotes the effective diffusion coefficient,

$$\mathscr{D}_{\mathrm{eff}}=\mathscr{D}_{12}+R_0^2\bar{U}^2/48\mathscr{D}_{12}. \qquad (3.2\text{–}15)$$

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

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

where $V=\pi R_0^2 L$ is the tube volume. This is a skewed, not Gaussian, distribution, but if $\mathscr{D}_{\mathrm{eff}}/\bar{U}L \leqslant 0.01$, the distribution becomes nearly Gaussian [34]. The reason is that $\bar{U}t/L$ must be nearly unity when $\mathscr{D}_{\mathrm{eff}}/\bar{U}L$ 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

$$\tau^2=\frac{2\mathscr{D}_{\mathrm{eff}}}{\bar{U}L}=\frac{2\mathscr{D}_{12}}{\bar{U}L}+\frac{R_0^2\bar{U}}{24\mathscr{D}_{12}}. \qquad (3.2\text{–}17)$$

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

$$w_{1/2}=2(2\ln 2)^{1/2}\tau. \qquad (3.2\text{–}18)$$

The calculation of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ when $\bar{U} \leqslant (48)^{1/2}\mathscr{D}_{12}/R_0$, and the other root when $\bar{U} > (48)^{1/2}\mathscr{D}_{12}/R_0$. An experimental check is that calculated values of the physical $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ this level of precision is degraded because of the quadratic relationship between $w_{1/2}$ and $\mathscr{D}_{12}$.

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 $\mathscr{D}_{\mathrm{eff}}/\bar{U}L \geqslant 0.01$.

(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 $\mathscr{D}_{12}$ by gas chromatography have been reported to be about 1 to 2 percent. The reliability of these results is best estimated by comparisons with $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$.

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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$, 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

$$J_1 = -n\mathscr{D}_{12}(\partial x_1/\partial z) + x_1(J_1 + J_2), \qquad (3.2\text{–}19)$$

$$J_2 = 0, \qquad (3.2\text{–}20)$$

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

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

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

$$(dL/dt) = J_1 M/N_0\rho_{\mathrm{liq}}, \qquad (3.2\text{–}22)$$

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 $\mathscr{D}_{12}$, 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 $\mathscr{D}_{12}$ 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	Altshuller 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	Grieveson 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 Kuvshinskiĭ	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
Schwertz 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 $\mathscr{D}_{12}$ is

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

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 $L^2/2\mathscr{D}_{12}$; to satisfy this condition experiments are usually run for several hours [39].

The calculated values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ are plotted as a function of the reciprocal of the observed length of the diffusion path L; the corrected values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ [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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ [40]. This shows that liquids should be degassed prior to their use.

(4) The equilibrium values used in the formulas for $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 temperatures the scatter in the data is even greater [44]. In another review of $\mathscr{D}_{12}$ 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, H_2-H_2O 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$. 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 evived and improved by Frost.

TABLE 8. *Determinations of $\mathscr{D}_{12}$ 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		
von Obermayer	1883, '87	Buckingham	1904
Toepler	1896	Wicke and Kallenbach	1941
Foch	1913	Weisz	1957
Barus	1924 a, b	Bendt [b]	1958
Currie	1960	Scott and Cox	1960
Frost	1967	Evans et al.	1961
Kaufmann	1967	Wicke and Hugo	1961
Rhodes and Amick	1967	Scott and Dullien	1962
Zhukhovitskiĭ et al	1968	Evans et al.	1962, '63
		Kosov and Kurlapov [b]	1966
b. Back Diffusion		Coates and Mian	1967
		Henry et al.	1967
Harteck and Schmidt	1933	Mian	1967
Van der Held and Miesowicz	1937	Reist	1967
Spier	1939, '40	Zhalgasov and Kosov [b]	1968
		Ellis and Holsen	1969
c. Capillary Leak		Mian et al.	1969
		Hawtin et al.	1969
Klibanova et al.	1942	Schneider and Schäfer	1969
Kosov	1957	Kosov and Zhalgasov [b]	1970
Vyshenskaya and Kosov	1959, '65		
Kosov and Karpushin	1966	f. Dissociated Gases	
De Paz et al.	1967		
		Krongelb and Strandberg	1959
d. Unsteady Evaporation		Wise	1959, '61
		Walker	1961
Mullaly and Jacques	1924	Young	1961
Mackenzie and Melville	1932, '33	Morgan and Schiff	1964
Arnold	1944	Yolles and Wise	1968
Fairbanks and Wilke	1950	Khouw et al.	1969
Nelson	1956	Sancier and Wise	1969
de Nordwall and Flowers	1958	Baker	1970 b
Nikolaev and Aleskovskiĭ	1964	Yolles et al.	1970
Petit	1965		

[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 $\mathscr{D}_{12}$, 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 $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ up to 882 K. These studies require the calibration of the porosity of the system by means of independently published values of $\mathscr{D}_{12}$.

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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ for dissociated gases which probably give more reliable results than the present direct methods. For example, $\mathscr{D}_{12}$ for H-H_2 can be obtained from measurements of the mixture viscosity [48], and $\mathscr{D}_{12}$ for N-N_2 and O-O_2 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 $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$, and results are available for about ten gas pairs at 293 K. These results have varied agreements, within 10 percent, with $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$. The results, however,

TABLE 9. *Determinations of* $\mathscr{D}_{12}$ *by miscellaneous experimental methods* [a]

Author(s)	Date
a. Droplet Evaporation	
Langmuir	1918
Topley and Whytlaw-Gray	1927
Houghton	1933
Bradley et al.	1946
Bradley and Shellard	1949
Birks and Bradley	1949
Bradley	1951
Bradley and Waghorn	1951
Katan [b]	1969
b. Dufour Effect	
Waldmann	1944, '47
Mason et al.	1967
c. Thermal Separation Rate	
Nettley	1954
van Itterbeek and Nihoul	1957
Lonsdale and Mason	1957
Saxena and Mason	1959
Weissman et al.	1961
Wendt et al.	1963
Mason et al.	1964 b
d. Kirkendall Effect	
McCarty and Mason	1960
Mason	1961
e. Sound Absorption	
Holmes and Tempest	1960
Carey et al.	1966
Carey et al.	1968
f. Cataphoresis	
Freudenthal	1966
Hogervorst and Freudenthal	1967
Hogervorst	1971
g. Resonance Methods	
(Nuclear Magnetic Resonance)	
Luszczynski et al.	1962
Lipsicas	1962
Hartland and Lipsicas	1963
Luszczynski et al.	1967
Khoury and Kobayashi	1970
(Optical Pumping) [c]	
Franzen	1959
Bernheim	1962
McNeal	1962
Anderson and Ramsey	1963
Legowski	1964
Ramsey and Anderson	1964
Bernheim and Korte	1965
Gozzini et al.	1967
(Mercury Band Fluorescence)	
Coulliette	1928
Biondi	1953
McCoubrey	1954
McCoubrey and Matland	1954
Matland and McCoubrey	1955
McCoubrey and Matland	1956
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 bead 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 $\mathscr{D}_{12}$. The marker is located in a tube connected in parallel to a two-bulb apparatus. A value of $\mathscr{D}_{12}$ has been obtained for He-Ar at 303 K, which is in excellent agreement with directly determined $\mathscr{D}_{12}$.

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 $\mathscr{D}_{12}$, 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 $\mathscr{D}_{12}$, 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 $\mathscr{D}_{12}$ 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 fluoresence). The names in parentheses indicate the groupings for the studies listed in table 9. The nuclear magnetic resonance technique has been used to determine $\mathscr{D}_{12}$ 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 ^{3}He. The optical pumping technique has been used to determine $\mathscr{D}_{12}$ of alkali metal vapors (Na, Rb, and Cs) in various other gases.

References for Section 3

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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 $\mathscr{D}_{12}$.

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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ from direct measurements included the following factors:

(1) experimental method,

(2) reproducibility of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$, 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 apparent that the small composition dependence of $\mathscr{D}_{12}$ had to be taken into account, and that compact means of summarizing values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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,

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

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

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

$$b = (-S_2/Q_2)(Q_1/S_1) - 1. \qquad (4.2\text{–}3)$$

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

$$b + 1 = 10a(Q_1/P_1). \qquad (4.2\text{–}4)$$

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

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

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

$$b + 1 = 10a(1 + 1.8m + 3m^2), \qquad (4.2\text{–}6)$$

where

$$m = M_2/M_1 < 1. \qquad (4.2\text{–}7)$$

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

$$a \approx \frac{2^{1/2}}{8(1+1.8m)^2}\,\frac{\overline{\Omega}_{12}^{(1,1)}}{\overline{\Omega}_{22}^{(2,2)}}. \qquad (4.2\text{–}8)$$

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 $\mathscr{D}_{12}$ and η_2, the viscosity of the light component, whereby eq (4.2–8) may be written as

$$a \approx \frac{3(1+m)^{1/2}}{20(1+1.8m)^2}\,\frac{\eta_2 RT}{p\mathscr{D}_{12}M_2}. \qquad (4.2\text{–}9)$$

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 $\mathscr{D}_{12}$ 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,

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

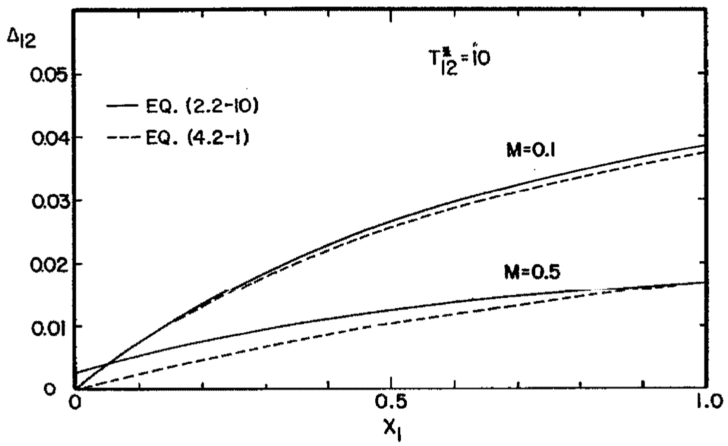

FIGURE 3. *Composition dependence of* Δ_{12} *at two mass ratios* (m).

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 $\mathscr{D}_{12}$ can be easily accommodated by adjustment of values of ζ.

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

$$\mathscr{D}_{12}(x_1=1/2) = \mathscr{D}_{12}(x_1)\left[\frac{1+\Delta_{12}(x_1=1/2)}{1+\Delta_{12}(x_1)}\right], \qquad (4.2\text{–}10)$$

where $\mathscr{D}_{12}(x_1)$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $p\mathscr{D}_{12}$. Over a narrow range of temperatures a plot of $\ln(p\mathscr{D}_{12})$ 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,

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

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 $\mathscr{D}_{12}$ 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

$$\ln\,(p\mathscr{D}_{12}) = \ln A + s\ln T - (S/T). \qquad (4.3\text{–}2)$$

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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ has the asymptotic (classical) form,

$$p\mathscr{D}_{12} = AT^{11/6}, \qquad T \rightarrow 0, \qquad (4.3\text{–}3)$$

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

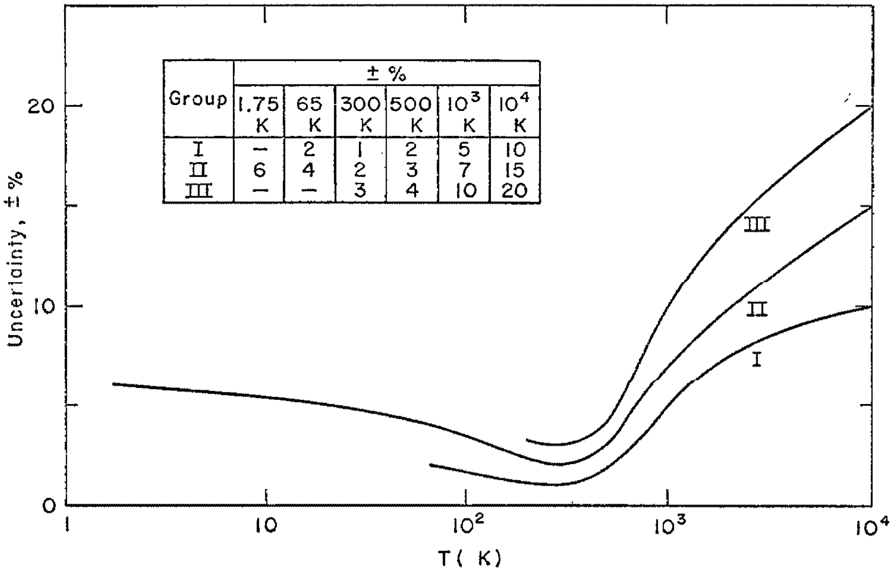
This chapter summarizes the most reliable experimental results for binary gaseous diffusion coefficients through 1968. Gas pairs for which only limited or uncertain data exist are not included here, but these experimental measurements may be retrieved with the aid of table 16, which extends through 1970.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

5.1. Uncertainty Limits

The sources of reliable values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ from direct experiment have been extended in both directions by the use of data on mixture viscosities, or, in a few instances, on thermal diffusion factors. These derived values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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).



Group	± %					
	1.75 K	65 K	300 K	500 K	10^3 K	10^4 K
I	–	2	1	2	5	10
II	6	4	2	3	7	15
III	–	–	3	4	10	20

FIGURE 4. *Estimated uncertainty limits of $\mathscr{D}_{12}$ as a function of temperature.*

TABLE 10. *Grouping of systems according to uncertainty limits of $\mathscr{D}_{12}$.*

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_2O,SF_6) SF_6-(He,Ar,H_2,CH_4,N_2,CO,O_2,air,CO_2)
Miscellaneous Systems	H_2O-(N_2,O_2,air,CO_2) CO_2-(Ne,H_2O,C_3H_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_2O-N_2	282 to 373	4
H_2O-O_2	282 to 1070	7
H_2O-air	282 to 1070	5 to 10
H_2O-CO_2	296 to 1640	10 to 7
CO_2-Ne	195 to 625	3 to 5
CO_2-C_3H_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 > 1000	15 30

The gas pairs of Group I have the most reliable values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 H_2-N_2 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 H_2-N_2 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 $\mathscr{D}_{12}$ 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-O_2, H_2-Ne, and N_2-Ar, do not have as many reliable measurements as the other gas pairs of Group II. The fourth gas pair, H_2-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-N_2, or almost identical values of $\mathscr{D}_{12}$.

The gas pairs of Group III have a relatively small number of reliable measurements of $\mathscr{D}_{12}$ at about room temperature. At temperatures above 1000 K the values of $\mathscr{D}_{12}$ have relatively large uncertainties because large discrepancies exist in the molecular-beam measurements used to calculate values of $\mathscr{D}_{12}$, 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 H_2O-CO_2 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ for N_2 and O_2 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* $\mathscr{D}_{12}$

System	10^3A	s	$10^{-8}\frac{\varphi_0}{k}$	S	S'	T Range	Group
	$\frac{\text{atm-cm}^2}{\text{s(K)}^s}$		K	K	$(K)^2$	K	
^3He-^4He	32.4	1.501	0.0448	−0.9630	1.894	1.74–10^4	II
^3He-^4He	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	II
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* $\mathscr{D}_{12}$

System	10^5A	s	S	T Range	Group
	$\frac{\text{atm-cm}^2}{s(K)^s}$		K	K	
He-CH_4	3.13	1.750	—	298–10^4	III
He-O_2	4.37	1.710	—	244–10^4	II
He-air	3.78	1.729	—	244–10^4	II
He-CO_2	3.31	1.720	—	200–530	II
He-SF_6	3.87	1.627	—	290–10^4	III
Ne-H_2	5.95	1.731	—	90–10^4	II
Ne-N_2	1.59	1.743	—	293–10^4	III
Ne-CO_2	1.07	1.776	—	195–625	Misc.
Ar-CH_4	0.784	1.785	—	307-10^4	III
Ar-N_2	.904	1.752	—	244–10^4	II
Ar-CO	.904	1.752	—	244–10^4	III
Ar-O_2	.977	1.736	—	243–10^4	III
Ar-air	.917	1.749	—	244–10^4	III
Ar-CO_2	1.74	1.646	89.1	276–1800	III
Ar-SF_6	1.48	1.596	145.4	328–10^4	III
Kr-N_2	0.653	1.766	—	248–10^4	III
Kr-CO	.653	1.766	—	248–10^4	III
Xe-H_2	3.68	1.712	16.9	242–10^4	III
Xe-N_2	0.470	1.789	—	242–10^4	III
H_2-CH_4	3.13	1.765	—	293–10^4	III
H_2-O_2	4.17	1.732	—	252–10^4	III
H_2-air	3.64	1.750	—	252–10^4	II
H_2-CO_2	3.14	1.750	11.7	200–550	II
H_2-SF_6	7.82	1.570	102.3	298–10^4	III
CH_4-N_2	1.00	1.750	—	298–10^4	III
CH_4-O_2	1.68	1.695	44.2	294–10^4	III
CH_4-air	1.03	1.747	—	298–10^4	III
CH_4-SF_6	1.10	1.657	69.2	298–10^4	III
N_2-O_2	1.13	1.724	—	285–10^4	III
N_2-H_2O	0.187	2.072	—	282–373	Misc.
N_2-CO_2	3.15	1.570	113.6	288–1800	II
N_2-SF_6	1.66	1.590	119.4	328–10^4	III
CO-O_2	1.13	1.724	—	285–10^4	III
CO-air	1.12	1.730	—	285–10^4	III
CO-CO_2	0.577	1.803	—	282–473	III
CO-SF_6	1.76	1.584	139.4	297–10^4	III
O_2-H_2O	0.189	2.072	—	282–450	Misc.
	2.78	1.632	—	450–1070	Misc.
O_2-CO_2	1.56	1.661	61.3	287–1083	III
O_2-SF_6	2.65	1.522	129.0	297–10^4	III
air-H_2O	0.187	2.072	—	282–450	Misc.
	2.75	1.632	—	450–1070	Misc.
air-CO_2	2.70	1.590	102.1	280–1800	III
air-SF_6	1.83	1.576	121.1	328–10^4	III
H_2O-CO_2	9.24	1.500	307.9	296–1640	Misc.
CO_2-N_2O	0.281	1.866	—	195–550	III
CO_2-C_3H_8	.177	1.896	—	298–550	Misc.
CO_2-SF_6	.140	1.886	—	328–472	III
H-He	14.2	1.732	—	275–10^4	Misc.
H-Ar	1.45	1.597	—	275–10^4	Misc.
H-H_2	11.3	1.728	—	190–10^4	Misc.
N-N_2	1.32	1.774	—	280–10^4	Misc.
O-He	4.68	1.749	—	280–10^4	Misc.
O-Ar	0.751	1.841	—	280–10^4	Misc.
O-N_2	1.32	1.774	—	280–10^4	Misc.
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 $\mathscr{D}_{12}$ based on molecular-beam measurements. Since the uncertainties in both cases are greater than the composition dependence of $\mathscr{D}_{12}$, 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 ^{3}He-^{4}He 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 $\mathscr{D}_{12}$ may be obtained from the classical asymptote, eq (4.3–3). The correlation constants for the classical asymptotes are given in table 14; a total of twenty-four gas pairs are listed which have London dispersion constants available.

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

System	$10^{6}A$	C[a]		ϵ/k[b]
	$\frac{\text{atm-cm}^2}{\text{s(K)}^{11/6}}$	$e^2a_0^5$	Λ^*[b]	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).

If estimates of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ at a given temperature. The larger value calculated is the better estimate of $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$ are required at pressures not equal to 1 atm, then the reciprocal pressure relationship of $\mathscr{D}_{12}$ is used, as discussed in section 2.2.

The values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$, 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 $\mathscr{D}_{12}$, 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 $\mathscr{D}_{12}$ as follows. First, the overall consistency of the data is rather good, although some reported values of $\mathscr{D}_{12}$ show considerable scatter. Second, careful appraisal of the experimental data is necessary to obtain the most reliable estimate of $\mathscr{D}_{12}$. A random selection of a value of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 uncentainty less than 1 percent.

TABLE 15. *Correlation parameters for the composition dependence of $\mathscr{D}_{12}$ according to eq (4.2–1)*

System	ζ	ϵ/k[a]	a	b	System	ζ	ϵ/k[a]	a	b
		K					K		
^3He-^4He	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_2O	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_2O	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_2O	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_2O-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 H_2-N_2 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 $\mathscr{D}_{12}$ 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 H_2-D_2. 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ and the usual "absolute" values (relative values of $\mathscr{D}_{12}$ are obtained by calibrating the apparatus against a mixture with known $\mathscr{D}_{12}$); (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 $\mathscr{D}_{12}$, for instance, mixture viscosity.

5.4. Detailed Remarks

This section presents the detailed remarks on the critical evaluation and the correlations of $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ used in the least-squares calculations of the reference equations, (2) intermolecular potentials obtained from molecular-beam experiments which were used to calculate $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ was disregarded at temperatures below 400 K for the ten noble gas pairs and H_2-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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ were taken to lie between these two extrapolations. At 10 000 K, the selected values approximate the mean of $\mathscr{D}_{12}$ 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 the calculated deviations would have exceeded the uncertainty limits of Group I. An additional point at 77 K ($\mathscr{D}_{12}=0.0607$) was used in the least-squares calculations, and it was weighted 3/10. This value of $\mathscr{D}_{12}$ 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 H_2-N_2 an additional datum at 562 K ($\log_{10} T=2.750$, $\log_{10}\mathscr{D}_{12}=0.365$) 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 $\mathscr{D}_{12}$ 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

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

System	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	
	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
H_2-N_2	.790	2.575

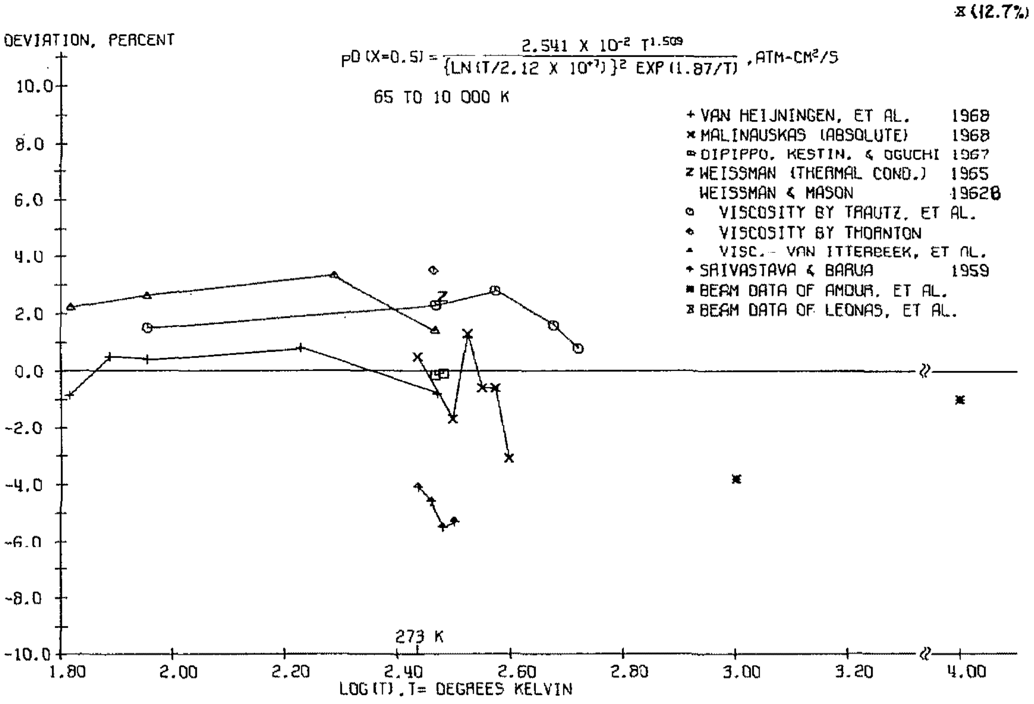


FIGURE 5. *Deviations of diffusion coefficients from reference equation.*

Helium—Neon

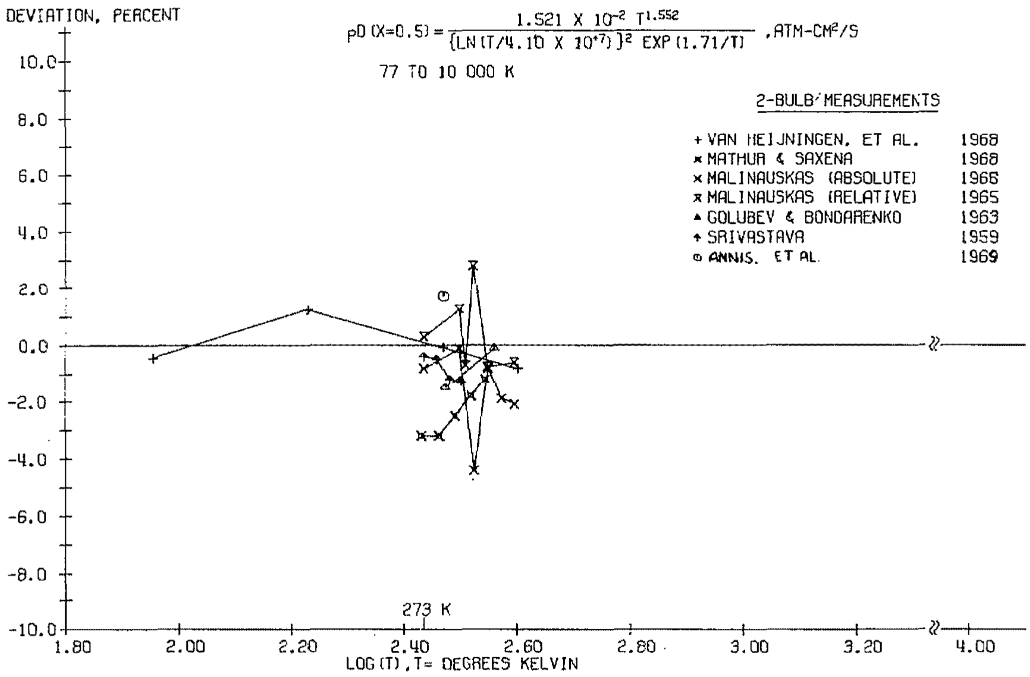


FIGURE 6. *Deviations of diffusion coefficients from reference equation.*

Helium—Argon

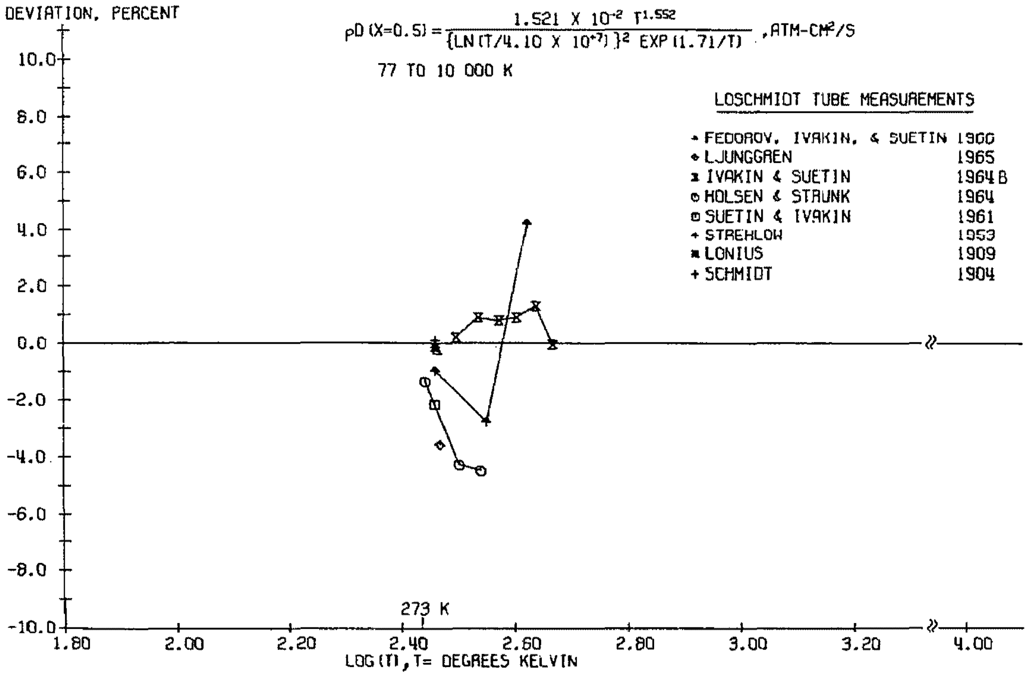


FIGURE 7. *Deviations of diffusion coefficients from reference equation.*

Helium—Argon

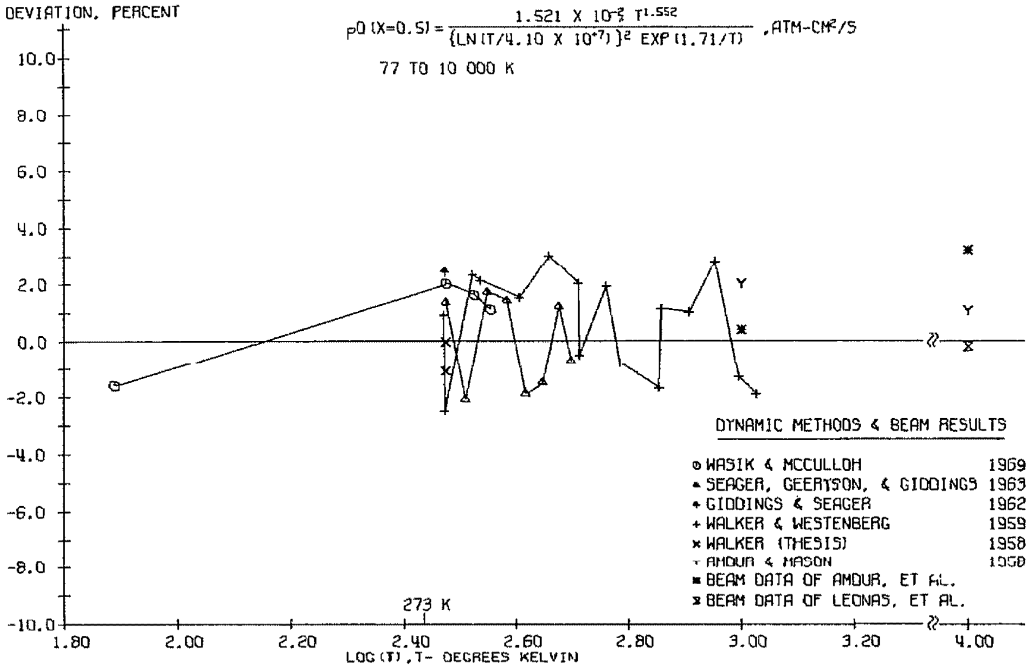


FIGURE 8. *Deviations of diffusion coefficients from reference equation.*

Helium—Argon

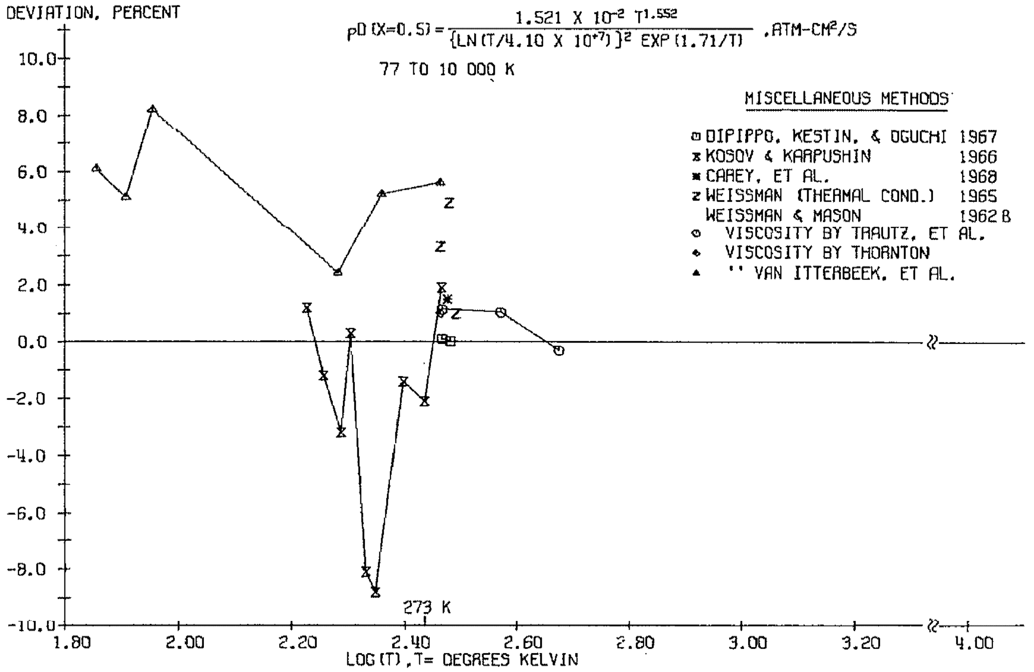


FIGURE 9. *Deviations of diffusion coefficients from reference equation.*

Helium—Argon

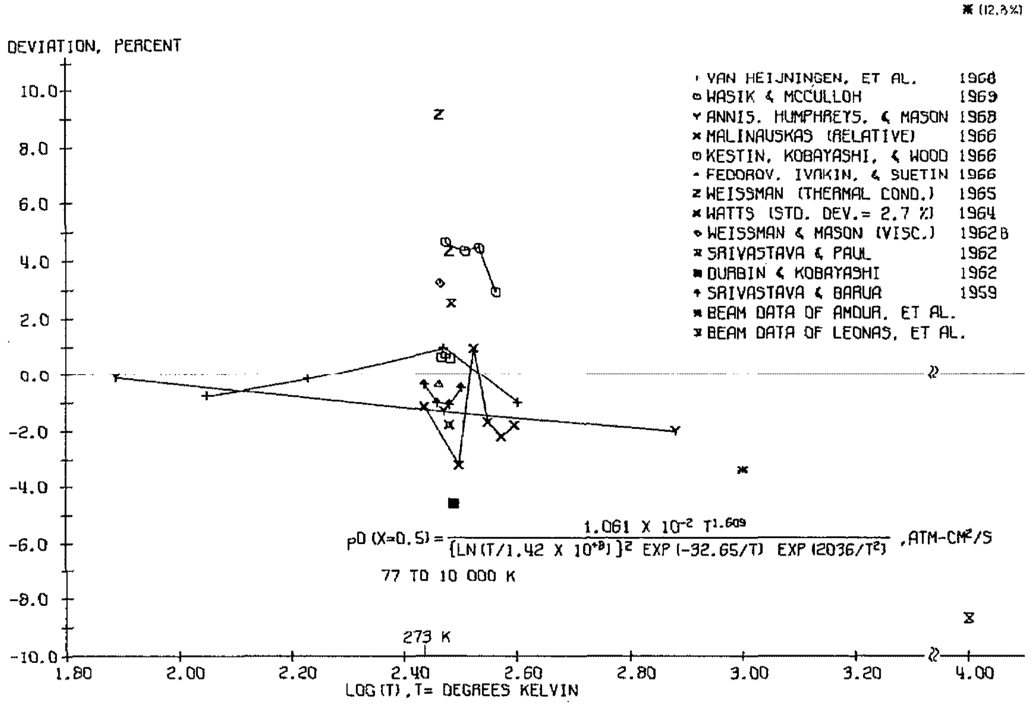


FIGURE 10. *Deviations of diffusion coefficients from reference equation.*

Helium—Krypton

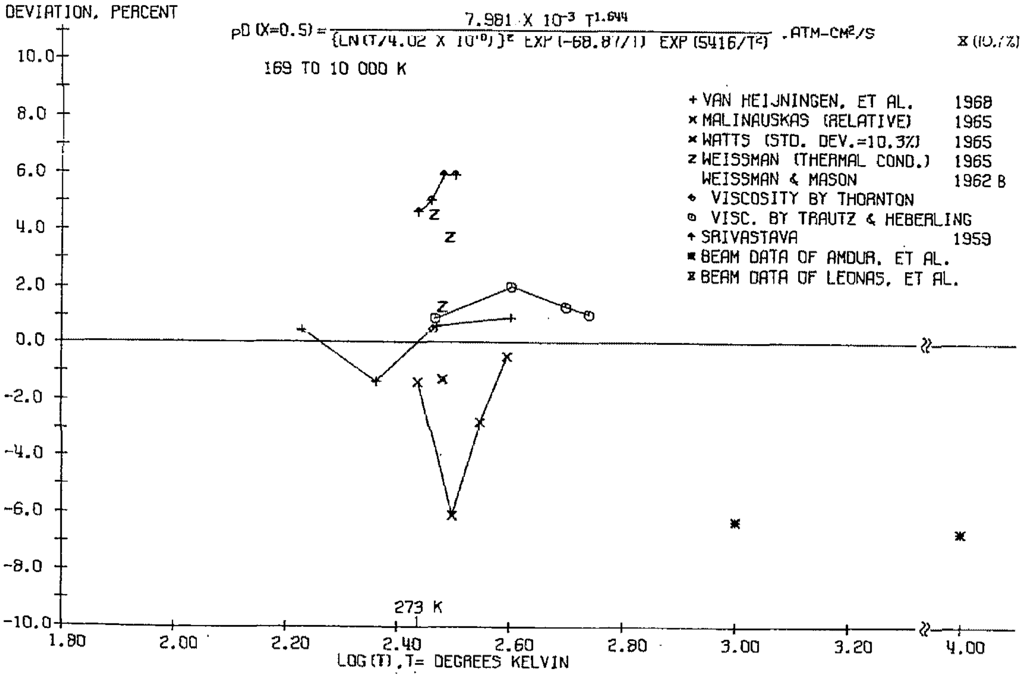


FIGURE 11. *Deviations of diffusion coefficients from reference equation.*

lelium—Xenon

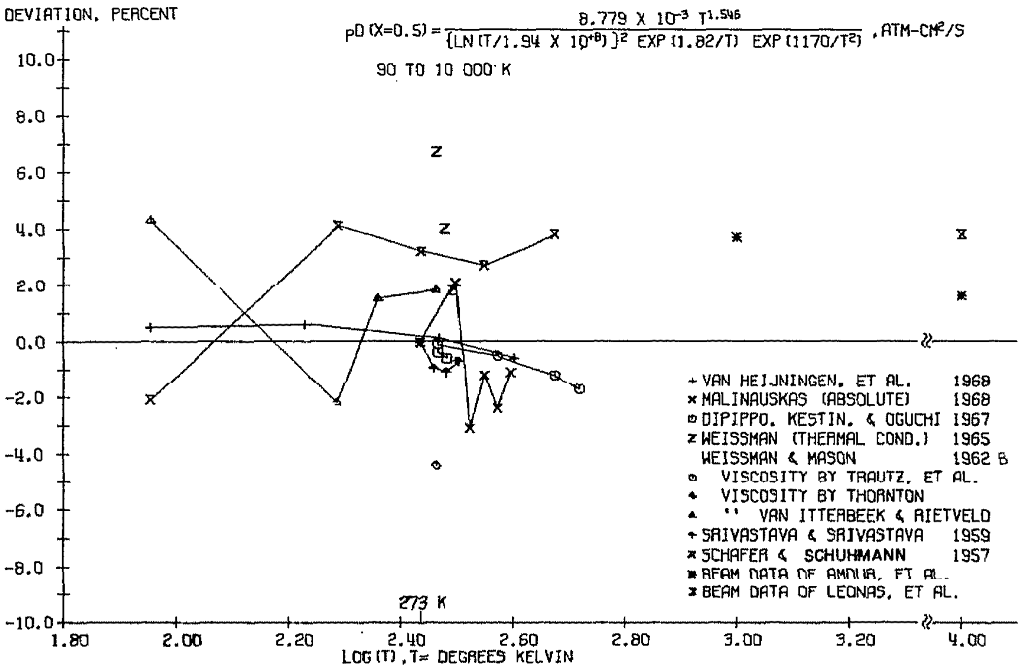


FIGURE 12. *Deviations of diffusion coefficients from reference equation.*

Neon—Argon

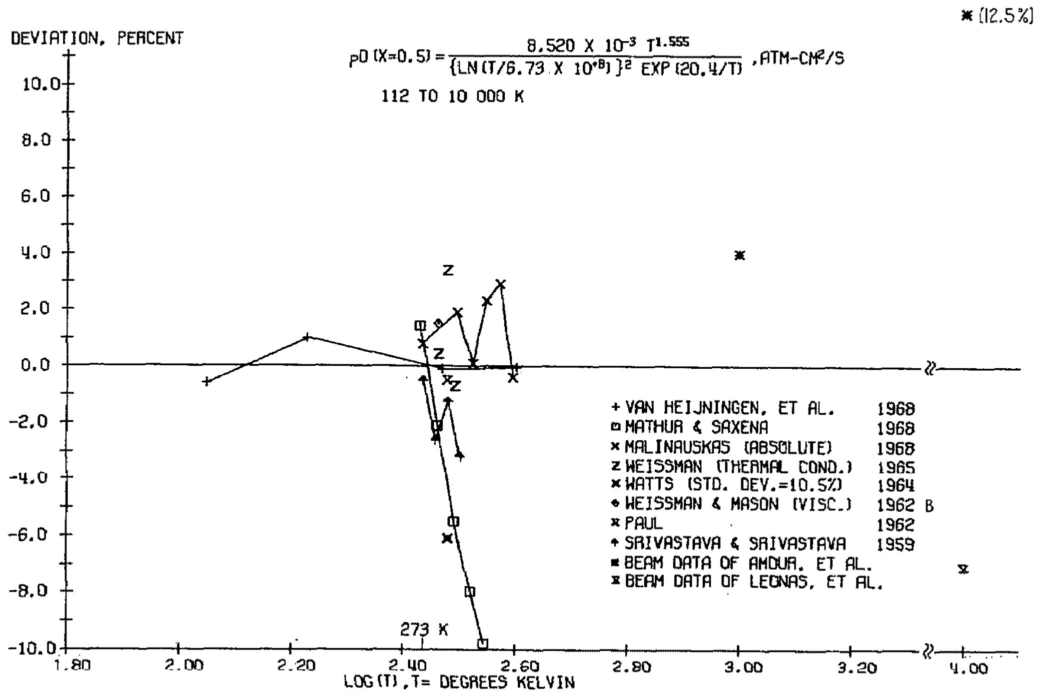


FIGURE 13. *Deviations of diffusion coefficients from reference equation.*

Neon—Krypton

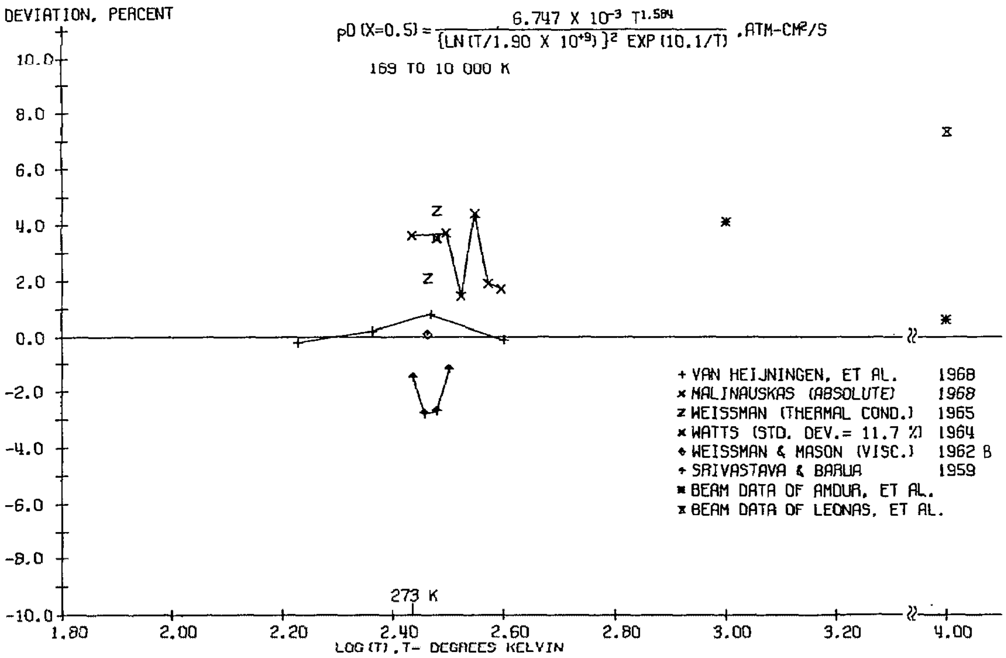


FIGURE 14. *Deviations of diffusion coefficients from reference equation.*

Neon—Xenon

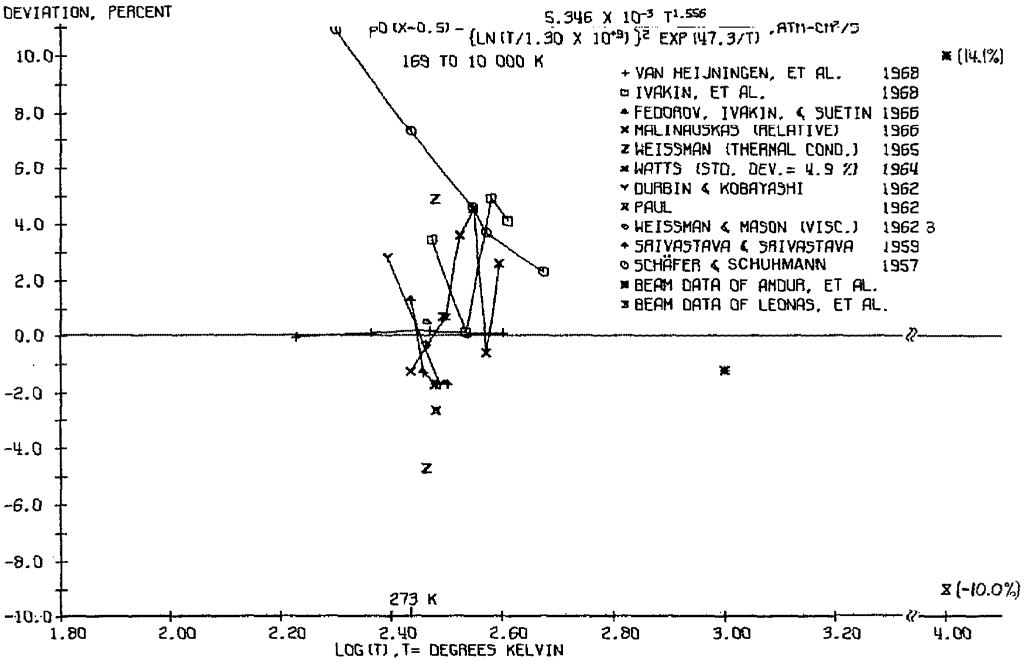


FIGURE 15. *Deviations of diffusion coefficients from reference equation.*

Argon—Krypton

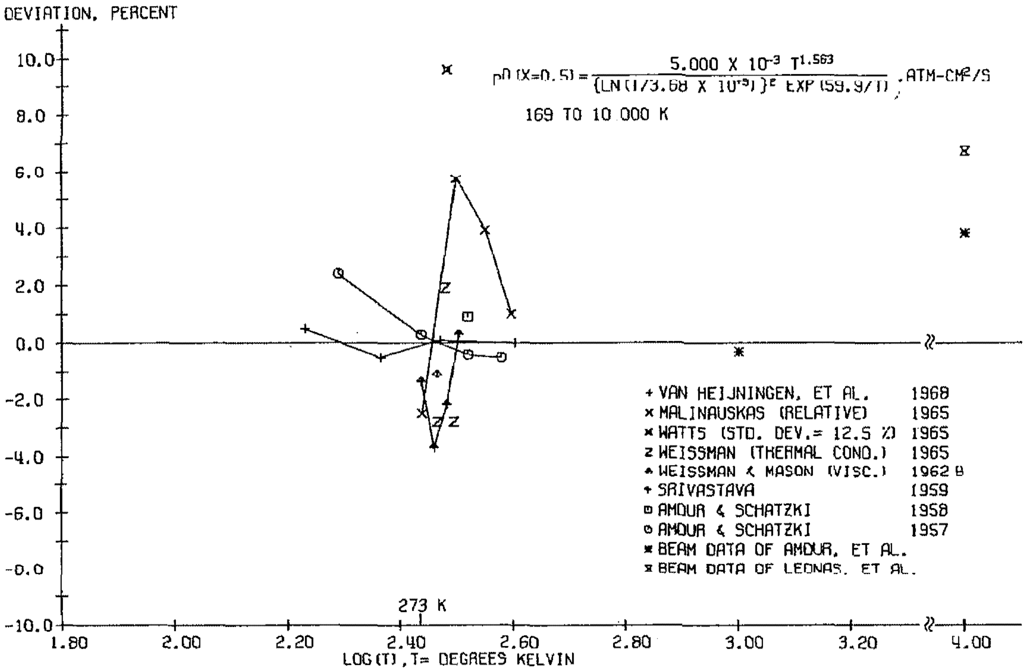


FIGURE 16. *Deviations of diffusion coefficients from reference equation.*

Argon—Xenon

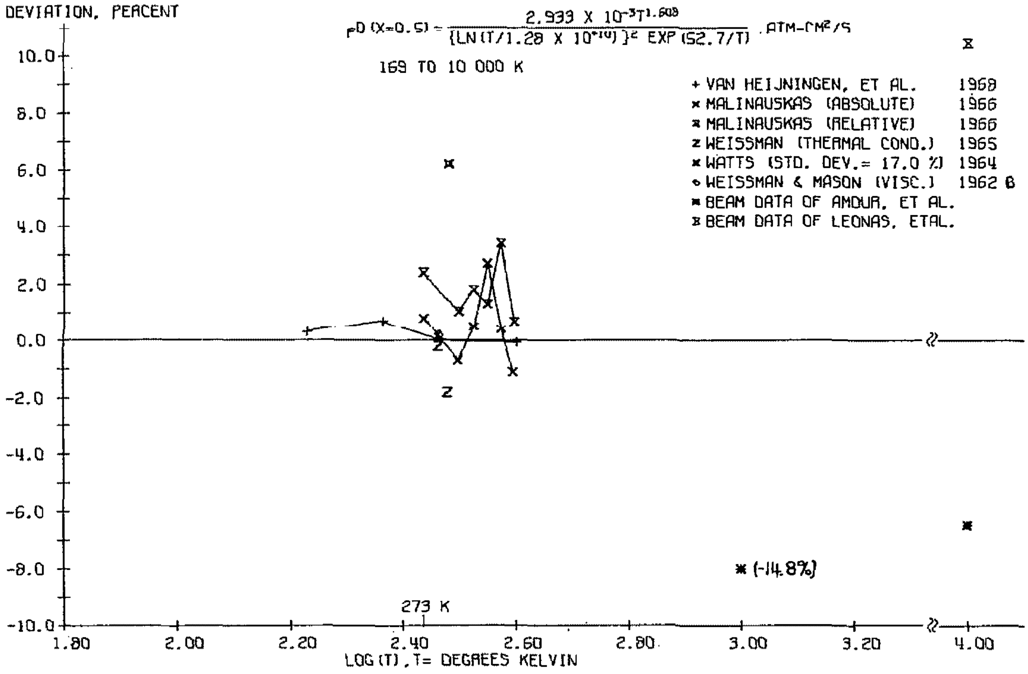


FIGURE 17. *Deviations of diffusion coefficients from reference equation.*

Krypton—Xenon

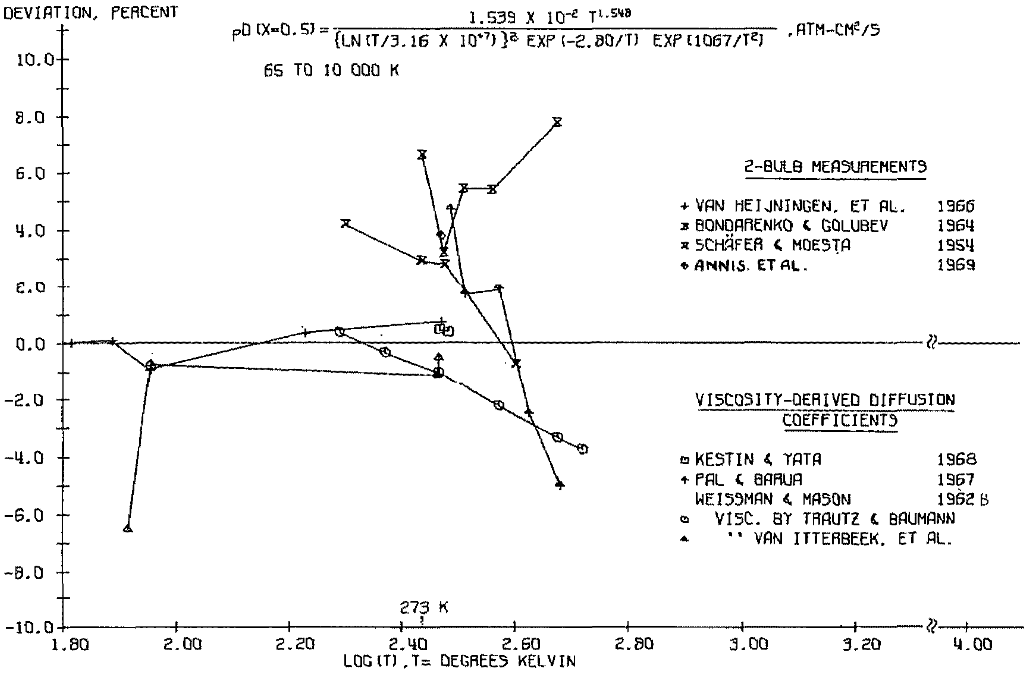


FIGURE 18. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Nitrogen

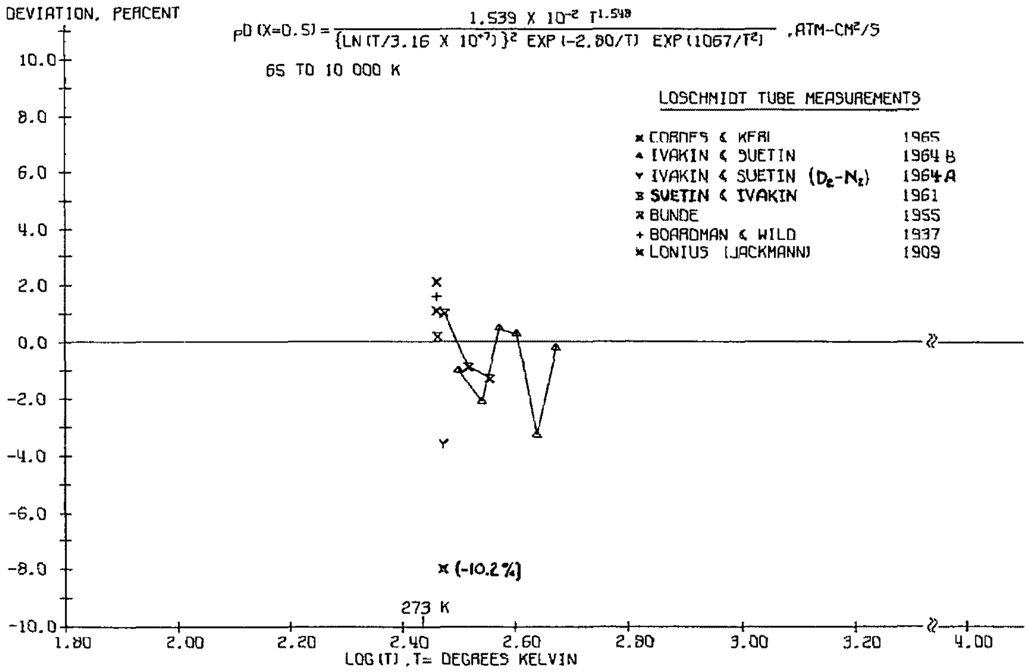


FIGURE 19. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Nitrogen

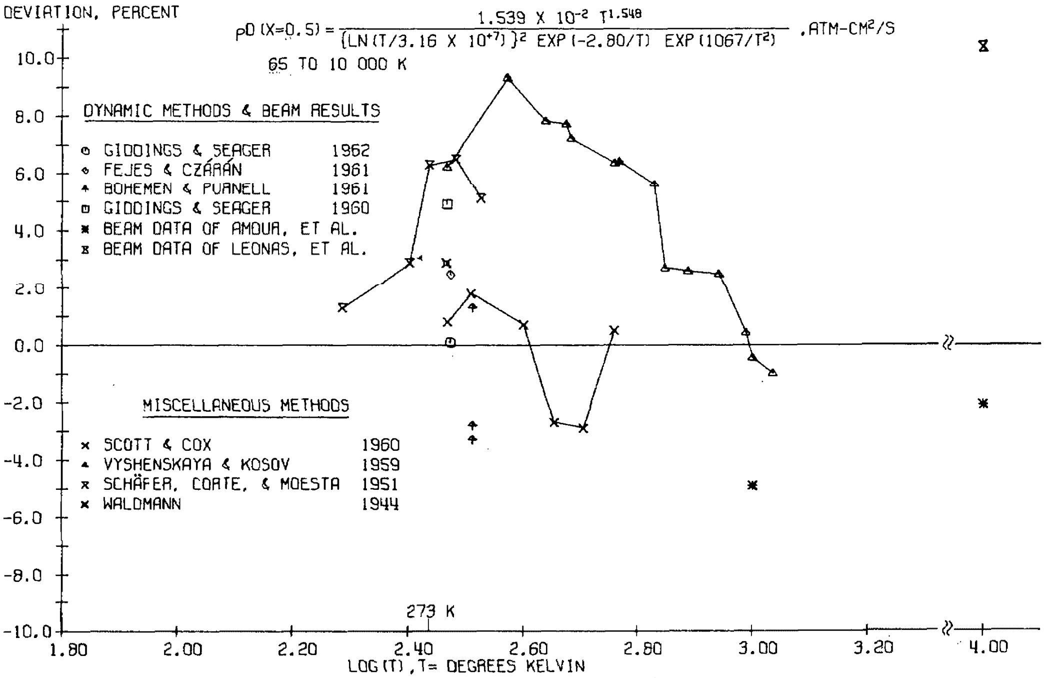


FIGURE 20. *Deviations of diffusion coefficients from reference equation.*

Hydrogen – Nitrogen

TABLE 18. *Molecular-beam potentials,* $\varphi(r) = K/r^s$, *for Group I* [a]

	Amdur et al.[b]			Leonas et al.[c]		
System	K, eV(Å)^s	s	Range, Å	K, eV(Å)^s	s	Range, Å
He-Ne	38.3	7.97	1.52–1.86	10.3	5.61	1.3 –1.65
He-Ar	62.1	7.25	1.64–2.27	22.6	5.15	1.63–2.06
He-Kr	27.4	5.68	1.85–2.37	45.3	5.52	1.67–2.04
He-Xe	182	6.96	2.14–2.60	35.2	5.2	1.73–2.2
Ne-Ar	630	9.18	1.91–2.44	99.5	6.56	1.93–2.49
Ne-Kr	223	7.71	2.09–2.64	437	7.65	2.15–2.52
Ne-Xe	1480	8.98	2.39–2.87	210	6.76	2.0 –2.56
Ar-Kr	367	6.88	2.30–2.92	855	6.92	2.4 –3.1
Ar-Xe	2450	8.15	2.60–3.15	292	5.9	2.48–3.27
Kr-Xe	1060	6.70	2.72–3.37	875	7.1	2.44–3.0
H_2-N_2	191.0	7.19	1.96–2.46	88.1	6.63_5	1.84–2.50

[a] Complete reference information is given in Bibliography II.
[b] All results except He-Ar and Ne-Ar are calculated by combination rules.
[c] Only the potential for H_2-N_2 is calculated by combination rules.

potentials for H_2-H_2 and N_2-N_2 which lead to an H_2-N_2 potential by application of the combination rules given in section 2.6, part c. Independent molecular-beam measurements have also been made by Amdur et al. for He-Ar, Ne-Ar, and the pairs He-He, Ne-Ne, Ar-Ar, Kr-Kr, and Xe-Xe. By application of the combination rules, this information also yielded potentials for all the noble gas pairs. Amdur et al. also measured potentials for He-H_2 and He-N_2, from which the H_2-N_2 potential was obtained. In table 18 the potential functions are listed which were used to calculate the deviation points shown on figures 5 to 20.

Special Comments. The lower temperature limits for He-Ne and Ne-Ar might have been extended to temperatures beyond the results established by van Heijningen et al. For He-Ne a datum was available at 20.4 K, and for Ne-Ar a datum at 65 K; both values of $\mathscr{D}_{12}$ are based on mixture viscosity (Weissman and Mason, 1962 b). After consideration of the reliability of the viscosity data, of A^*_{12}, and of the need for a quantum correction, these values of $\mathscr{D}_{12}$ were not used to extend the lower temperature limits of the reference equations for these systems.

The systems Kr-Xe and H_2-N_2 are borderline because of the greater uncertainties in their molecular-beam potentials.

In two-bulb measurements for noble gas pairs there are apparent systematic errors in results by three independent investigators:

(1) van Heijningen et al. (1968),
(2) Malinauskas (1965, 1966, 1968), and
(3) Srivastava (1959), Srivastava and Barua (1959), and Srivastava and Srivastava (1959).

The magnitudes of the errors are usually a few percent or less, and are discussed relative to the more accurate work by van Heijningen et al. The results by Malinauskas are slightly lower for the lighter gas pairs (He-Ne, He-Ar, He-Kr, He-Xe, and Ne-Ar) and higher for the heavier gas pairs (Ne-Kr, Ne-Xe, Ar-Kr, Ar-Xe, and Kr-Xe). A cause for this trend 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 $\mathscr{D}_{12}$ by direct measurements are in reasonable agreement with $\mathscr{D}_{12}$ determined from other transport properties, as shown in table 19. Here direct measurements by van Heijningen et al. (1966, 1968) are compared with $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ are really less reliable than the direct measurements of $\mathscr{D}_{12}$ because the uncertainties in the A^*_{12} values are no less than 1 percent. The other mixture-viscosity sources yield $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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-CO_2, H_2-air, H_2-CO_2 and N_2-CO_2 because when this work was done, there were no molecular-beam measurements available with air or CO_2.

The potentials by Amdur et al. for He-CO, He-O_2, and H_2-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 ^{3}He-^{4}He was taken the same as for ^{4}He-^{4}He, and that for H_2-D_2 the same as for H_2-H_2; 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.

^{3}He-^{4}He. This gas pair is exceptional because its assignment into Group II is based mainly on the reliability of values of $\mathscr{D}_{12}$ calculated from viscosity measurements. In some instances, viscosity data for ^{4}He-^{4}He (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 $\mathscr{D}_{12}$ were calculated from the ^{4}He viscosity data by Kalelkar and Kestin

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

TABLE 19. *Values of $\mathscr{D}_{12}$ 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	-0.1_5	+2.3	+3.5	+1.4	+2.6	+18.8
He-Ar	-0.0_5	+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	$+0.0_5$	—	—	+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 [a] (3.8)
Index of reliability	1	1	3	4	6	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	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Weight	Note
^{3}He-^{4}He	2.64	−3.1325	1/4	a
	4.15	−2.8125	1/4	a
	1.74	−3.4789	1/4	b
	2.00	−3.3665	1/4	b
	2.31	−3.2396	1/4	b
	2.66	−3.1355	1/4	b
	3.08	−3.0306	1/4	b
	3.96	−2.8386	1/4	b
	14.4	−1.9066	1	b
	19.6	−1.7012	1	b
	64.8	−0.8327	1	b
	76.1	−.7282	1	b
	192	−.0742	1	b
	296	.2253	1	b
	298.15	.2550	1	c
	373.15	.4214	1	c
	473.15	.5977	1	c
	573.15	.7405	1	c
	673.15	.8594	1	c
	773.15	.9614	1	c
	873.15	1.0527	1	c
	1010.15	1.1550	1	c
	1121.15	1.2307	1	c
	2039	1.7127	1/4	d
	7746	2.7774	1/4	d
	3377	2.1038	1/4	e
	10 000	2.9983	1/4	e
	2444	1.8639	1/4	f
	10 000	2.9908	1/4	f
	10 000	2.9895	1/4	g
He-N_2	77.2	−1.1331	2	h
	251	−0.265	1	*
	317	−0.100	1	*
	399	0.070	1	*
	501	.241	1	*
	631	.405	1	*
	794	.575	1	*
	1000	.745	1	*
	3170	1.640	2/5	*
	10 000	2.530	1/5	*
He-CO	—	—	—	i
He-O_2	317	−0.085	1	*
	10 000	2.480	1	*
He-air	282	−0.1818	1	j
	355	−.0119	1	j
	447	.1584	1	j
	1000	.7582	1	j
	10 000	2.4969	1	j
He-CO_2	200	−0.5229	1	k
	298.4	−.2240	1	l
H_2-He	90.1	−.7012	1	a
	194.7	−.1264	1	m
	251.2	.063	1	*
	317	.232	1	*
	399	.398	1	*
	501	.567	1	*
	1000	1.080	1/3	*
	3170	1.970	1/3	*
	10 000	2.900	1/6	*
H_2-Ne	90.1	−0.8416	1	a
	9505	2.6599	1	n
H_2-Ar	251.2	−0.220	1	*
	317	−.045	1	*
	399	.140	1	*
	501	.320	1	*
	631	.495	1	*
	794	.670	1	*
	1000	.845	1	*
	3170	1.710	1/3	*
	10 000	2.590	1/6	*
H_2-Kr	77.0	−1.270	1/5	*, l
	100	−1.040	1/5	*, l
	178	−0.563	1/5	*, l
	290.7	−.1688	1	o
	296.0	−.1564	1	p
	562	.334	1/5	*, l
	3160	1.632	1/5	*
	10 000	2.557	1/5	*
H_2-D_2	14.12	−2.3675	1	q
	15.47	−2.2832	1	q
	17.04	−2.1945	1	q
	18.70	−2.1051	1	q
	20.32	−2.0329	1	q
	90.0	−0.7721	1	q
	26.09	−1.8097	1	r
	32.57	−1.6091	1	r
	41.35	−1.4117	1	r
	48.06	−1.2832	1	r
	60.30	−1.1002	1	r
	70.32	−0.9851	1	r
	200.0	−.1925	1	s
	250.0	−.0292	1	s
	293.0	.0864	1	s
	400	.3181	1	s
	500	.4757	1	s
	763	.7882	1	s
	986	.9741	1	s
	3313	1.9047	1	t
	5000	2.2305	1	t
	10 000	2.7796	1	t
H_2-CO	—	—	—	u
H_2-air	282	−0.1487	1	v
	355	.0253	1	v
	447	.1987	1	v
	1000	.8048	1	v
	10 000	2.5635	1	v
H_2-CO_2	200.0	−0.5017	1	w
	298.15	−.1898	1	x
	473.0	.1673	1	y
N_2-Ar	316	−.664	1	*
	3160	1.088	1	*
N_2-CO	77.65	−1.7747	1/5	z
	194	−0.980	1/2	*
	251	−.790	1	*
	316	−.619	1	*
	398	−.450	1	*
	562	−.195	1/2	*
	1000	.226	1/5	*
	10 000	1.979	1/10	*
N_2-CO_2	298.15	−0.7825	1	x
	447	−.450	1/2	*
	708	−.095	1/2	*
	1000	.1553	1/2	aa
	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 $\mathscr{D}_{12}$ by Annis et al. (1969).
[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 H_2-N_2 is suitable because of isosteric molecules.
[v] Calculated from reference equations for H_2-N_2 and H_2-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 $\mathscr{D}_{12}$ 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	
^3He-^4He	4.71	5.94	1.27 –1.59	^4He-^4He	4.71	5.94	Amdur and Harkness (1954).
	4.33	5.86	1.10 –1.53	^4He-^4He	4.33	5.86	Belyaev and Leonas (1967 b).
He-N_2	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).
				N_2-N_2	550	7.4	Belyaev and Leonas (1966 a).
He-CO	40.3	5.91	1.55 –2.26	CO-Ar	551	6.99	Jordan et al. (1970).
				He-Ar	62.1	7.25	Amdur et al. (1954).
				Ar-Ar	849	8.33	Amdur and Mason (1954).
	92.24	7.045	1.705–2.225	He-He	4.33	5.86	Belyaev and Leonas (1967 b).
				CO-CO	1965	8.23	Belyaev et al. (1967).
He-O_2	19.5	7.26	1.47 –2.08	He-Ar	62.1	7.25	Amdur et al. (1954).
				Ar-O_2	1360	8.34	Jordan et al. (1970).
				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).
				O_2-O_2	240	6.3	Belyaev and Leonas (1967 a).
H_2-He	12.11	6.07	1.44 –1.76	Direct measurement			Amdur and Smith (1968).
	5.0	3.8	1.15 –1.89	Direct measurement			Belyaev and Leonas (1967 b).
H_2-Ne	98.55	8.095	1.685–2.03	He-He	4.71	5.94	Amdur and Harkness (1954).
				Ne-Ne	312	9.99	Amdur and Mason (1955 a).
				He-H_2	12.11	6.07	Amdur and Smith (1968).
	21	4.70	1.45 –2.215	He-H_2	5	3.8	Belyaev and Leonas (1967 b).
				Ne-Ne	78	7.65	Kamnev and Leonas (1965 a).
				He-He	4.33	5.86	Belyaev and Leonas (1967 b).
H_2-Ar	160	7.38	1.81 –2.44	He-H_2	12.11	6.07	Amdur and Smith (1968).
				He-Ar	62.1	7.25	Amdur et al. (1954).
				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-H_2	12.11	6.07	Amdur and Smith (1968).
				Kr-Kr	159	5.42	Amdur and Mason (1955 b).
				He-He	4.71	5.94	Amdur and Harkness (1954).
	89.33	4.72	1.80 –2.66	He-H_2	5	3.8	Belyaev and Leonas (1967 b).
				Kr-Kr	1382	7.7	Kamnev and Leonas (1966 a).
				He-He	4.33	5.86	Belyaev and Leonas (1967 b).
H_2-D_2	31.55	6.19	1.62 –1.96	He-H_2	12.11	6.07	Amdur and Smith (1968).
				He-D_2	12.27	6.06	Amdur and Smith (1968).
				He-He	4.71	5.94	Amdur and Harkness (1954).
	14.1	5.87	1.34 –1.95	H_2-H_2	14.1	5.87	Belyaev and Leonas (1967 b).
H_2-CO	107.4	5.81	1.89 –2.17	Ar-CO	551	6.99	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).
	166.4	7.05	1.82_5–2.43_5	H_2-H_2	14.1	5.87	Belyaev and Leonas (1967 b).
				CO-CO	1965	8.23	Belyaev et al. (1967).
N_2-Ar	755	7.78	2.28 –2.83	Direct measurement			Amdur et al. (1957).
	1050	8.16	2.12 –2.67	Direct measurement			Belyaev and Leonas (1967 a).
N_2-CO	2038	8.70	2.22 –2.77	Direct measurement			Belyaev et al. (1967).
	596	7.27	2.43 –3.07	N_2-N_2	596	7.27	Amdur et al. (1957).

[a] Potentials were not determined for air-(He,H_2) and CO_2-(He,H_2,N_2) because molecular-beam measurements were unavailable.

[b] Complete reference information is given in Bibliography II.

(1970). The direct measurements of $\mathscr{D}_{12}$ 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-N_2. 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 H_2-CO. The diffusion coefficients of He-CO and H_2-CO can be well approximated by those for He-N_2 and H_2-N_2 (Group I), respectively, because CO and N_2 are isosteric molecules and He-N_2 and H_2-N_2 have more reliable measurements than do He-CO and H_2-CO. A comparison of the reference equations with the reliable measurements by Ivakin and Suetin (1964 a, b) for He-CO and H_2-CO shows deviations less than about 2 percent. The H_2-CO system was not assigned to Group I, as is H_2-N_2, because of possible uncertainties due to the

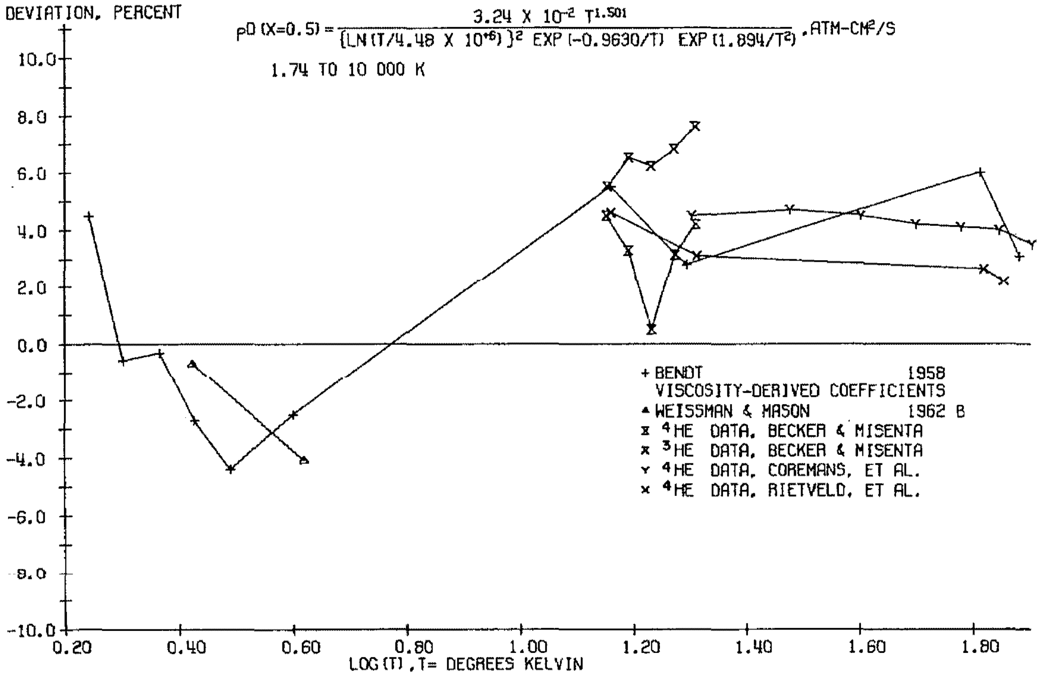

FIGURE 21. *Deviations of diffusion coefficients from reference equation.*

Helium-3—Helium-4

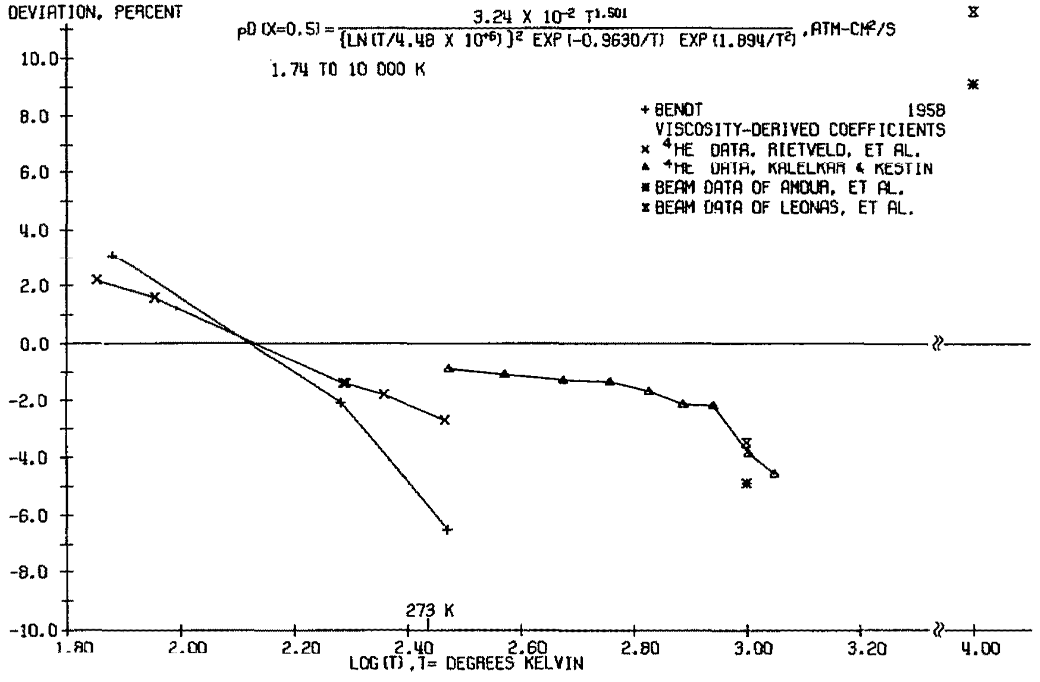

FIGURE 22. *Deviations of diffusion coefficients from reference equation.*

Helium-3—Helium-4

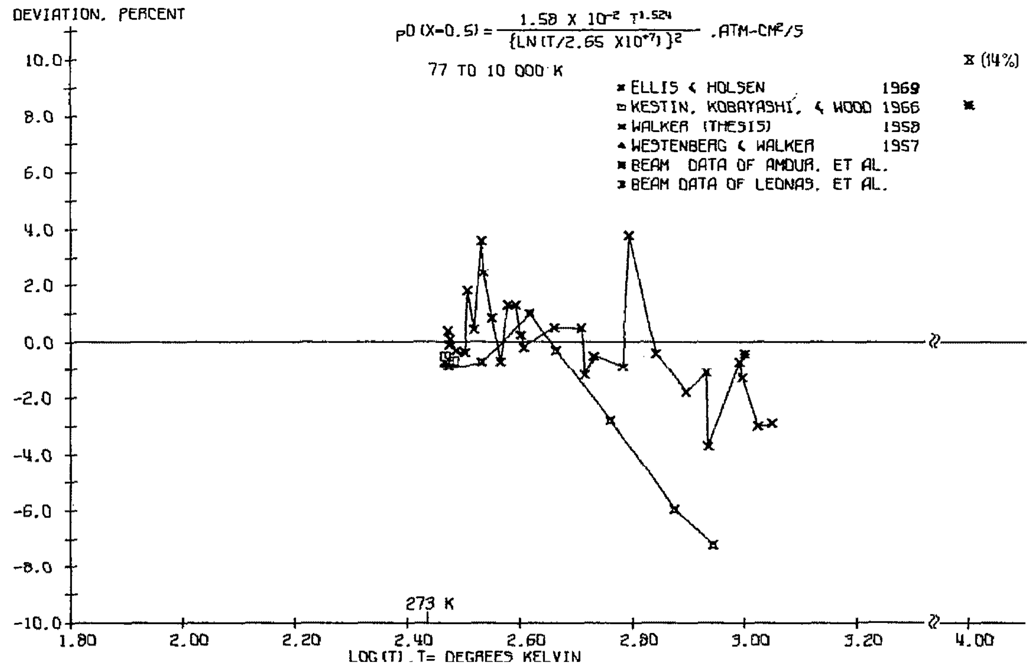

FIGURE 23. *Deviations of diffusion coefficients from reference equation.*

Helium—Nitrogen

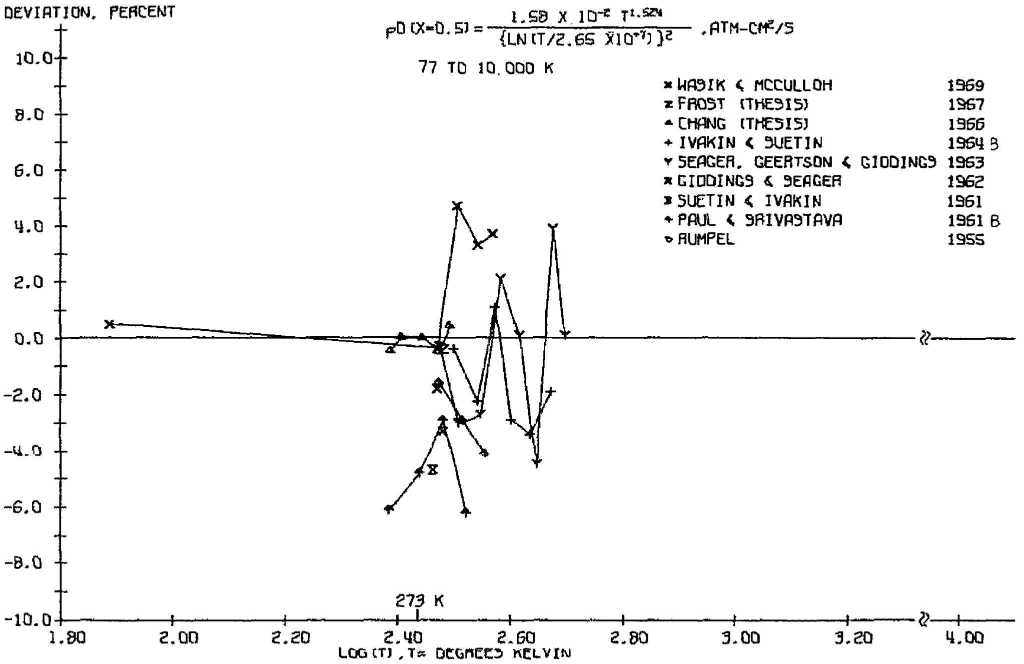

FIGURE 24. *Deviations of diffusion coefficients from reference equation.*

Helium—Nitrogen

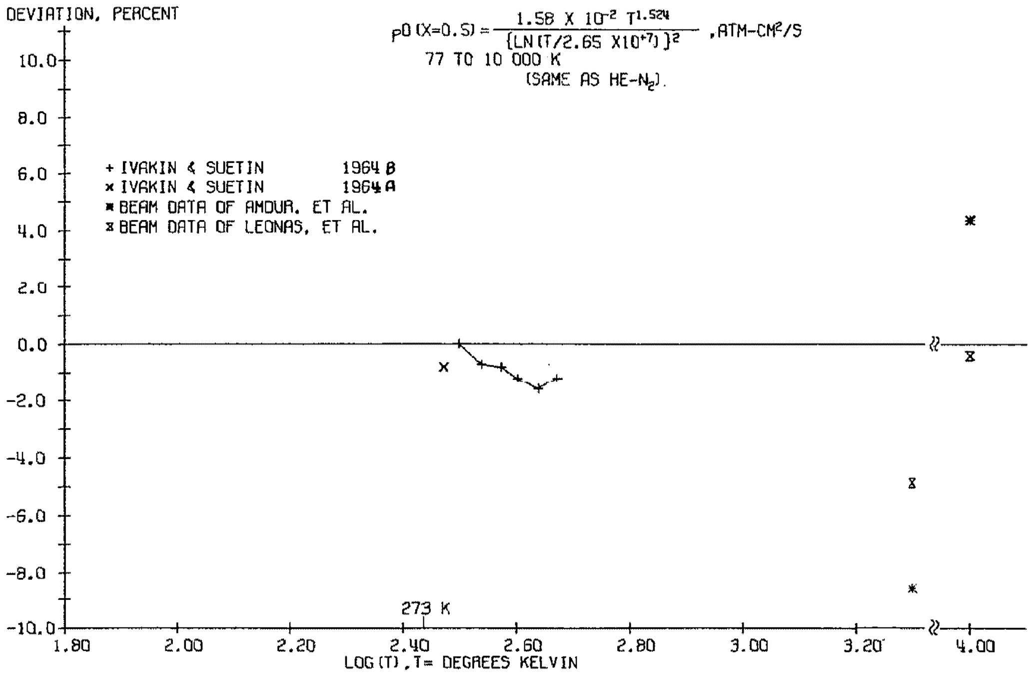

FIGURE 25. *Deviations of diffusion coefficients from reference equation.*

Helium—Carbon monoxide

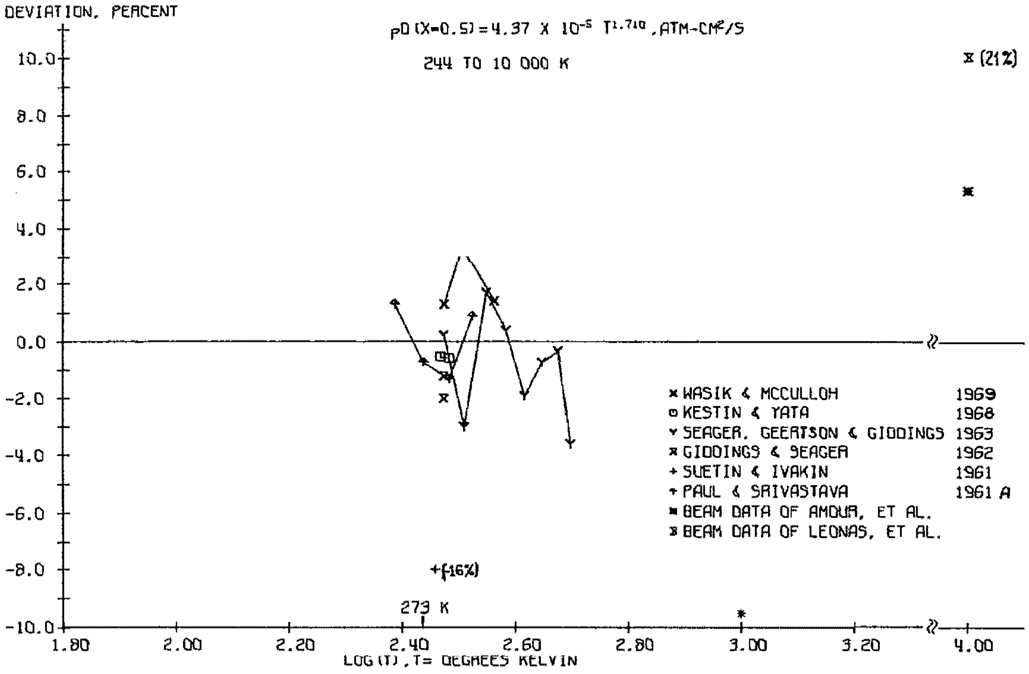

FIGURE 26. *Deviations of diffusion coefficients from reference equation.*

Helium—Oxygen

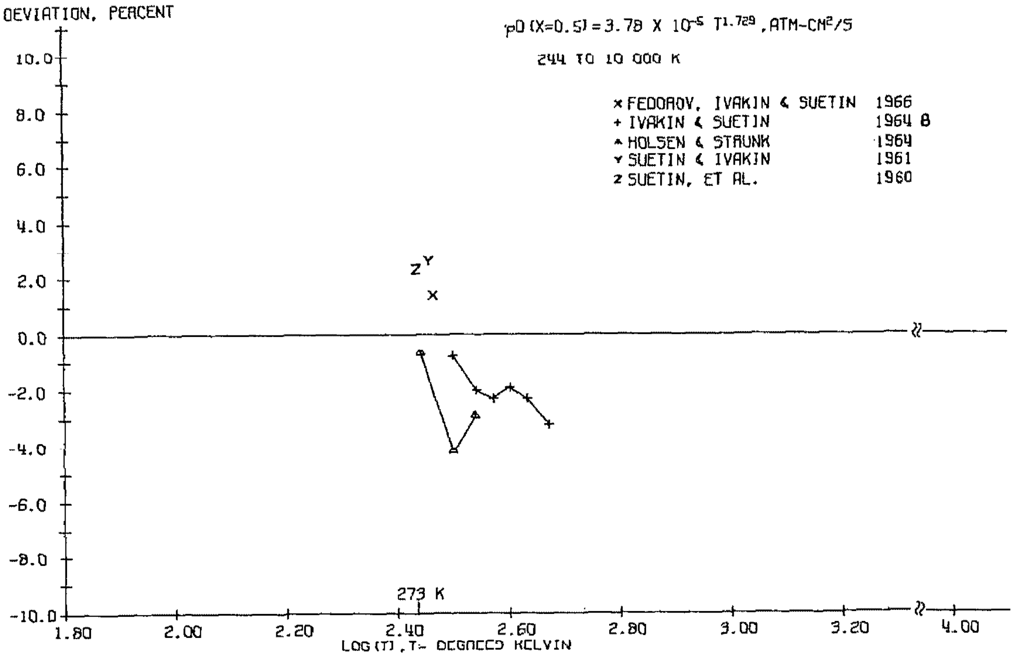

FIGURE 27. *Deviations of diffusion coefficients from reference equation.*

Helium—Air

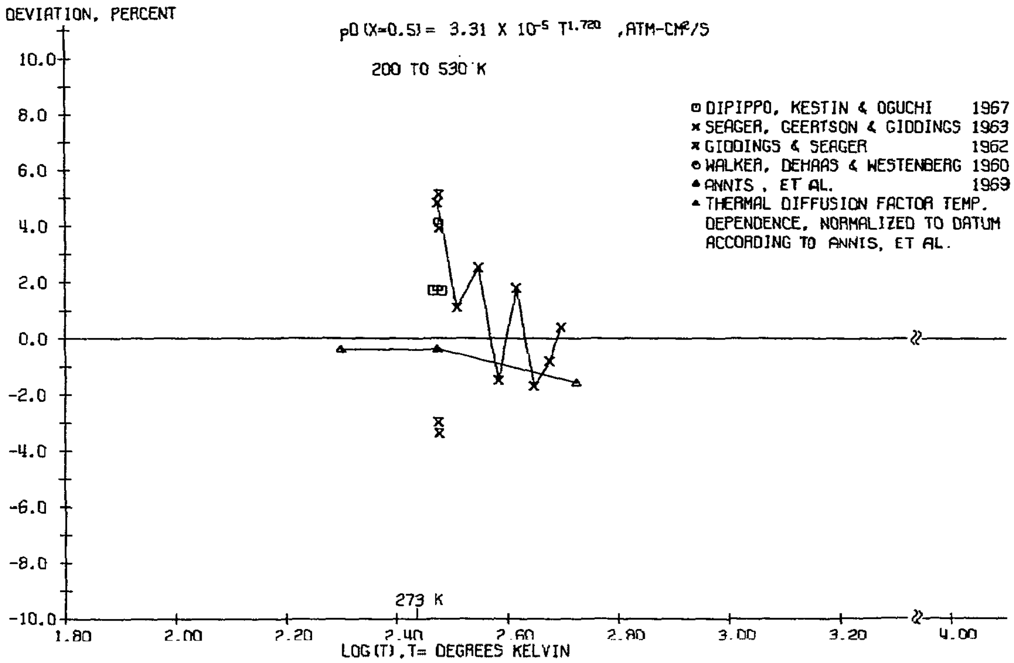

FIGURE 28. *Deviations of diffusion coefficients from reference equation.*

Helium—Carbon dioxide

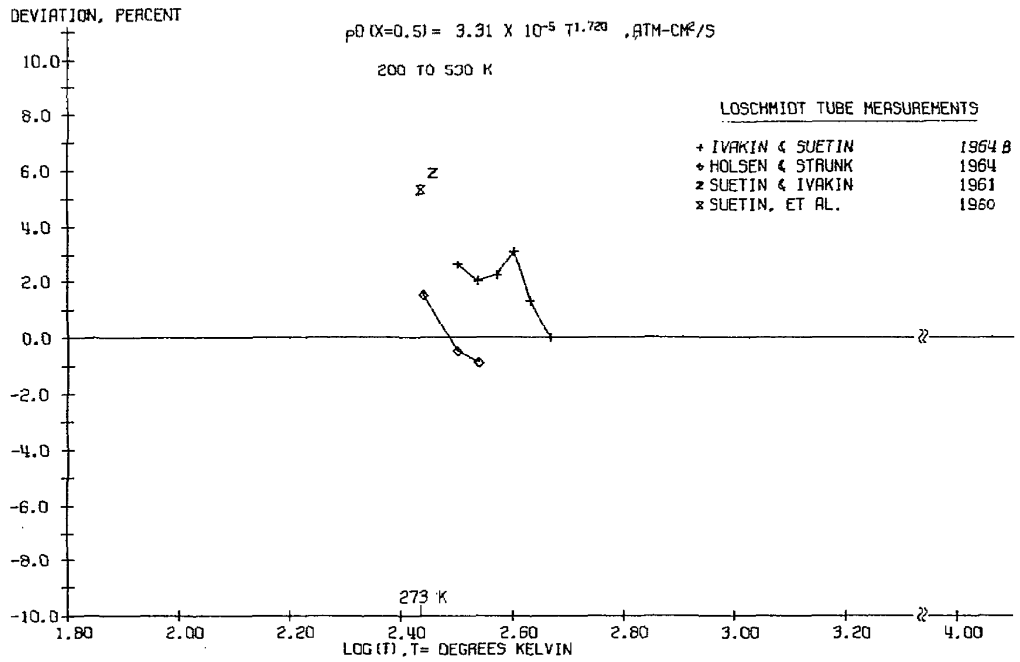

FIGURE 29. *Deviations of diffusion coefficients from reference equation.*

Helium – Carbon dioxide

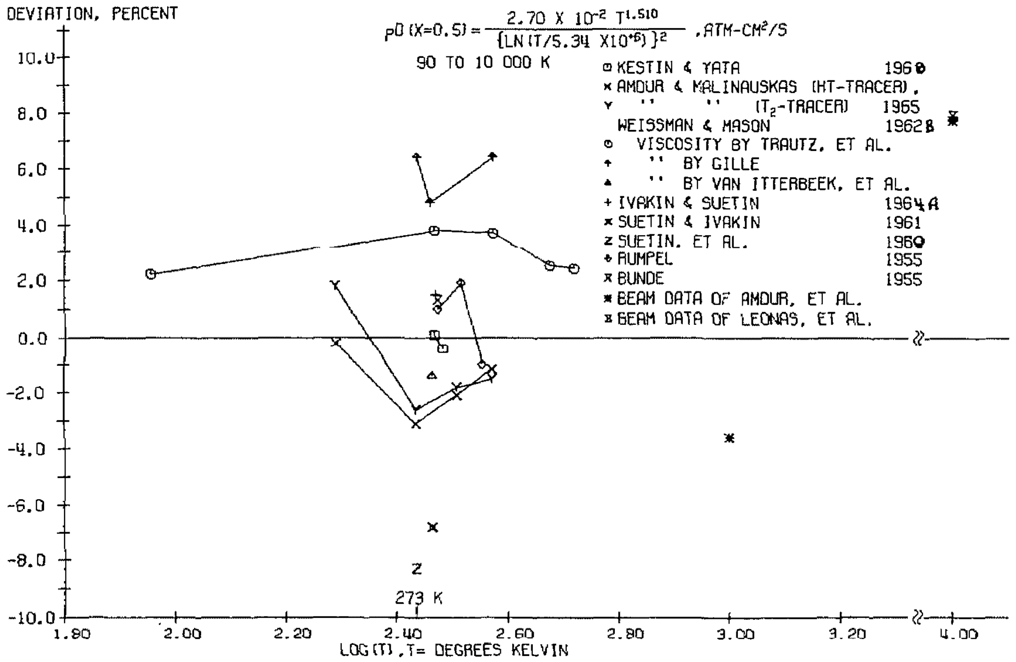

FIGURE 30. *Deviations of diffusion coefficients from reference equation.*

Hydrogen – Helium

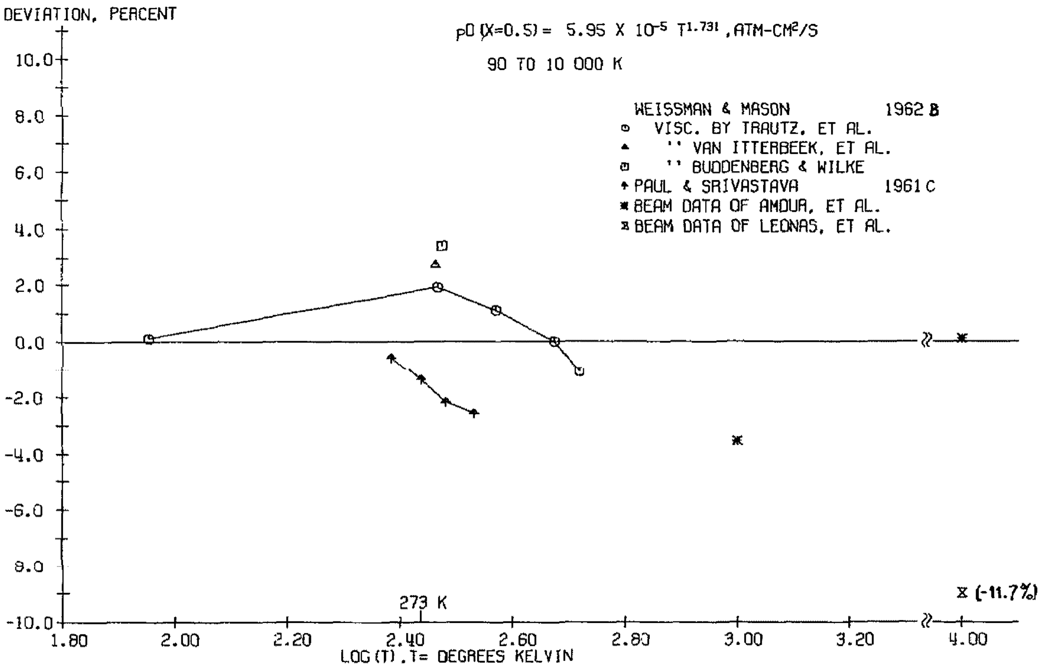

FIGURE 31. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Neon

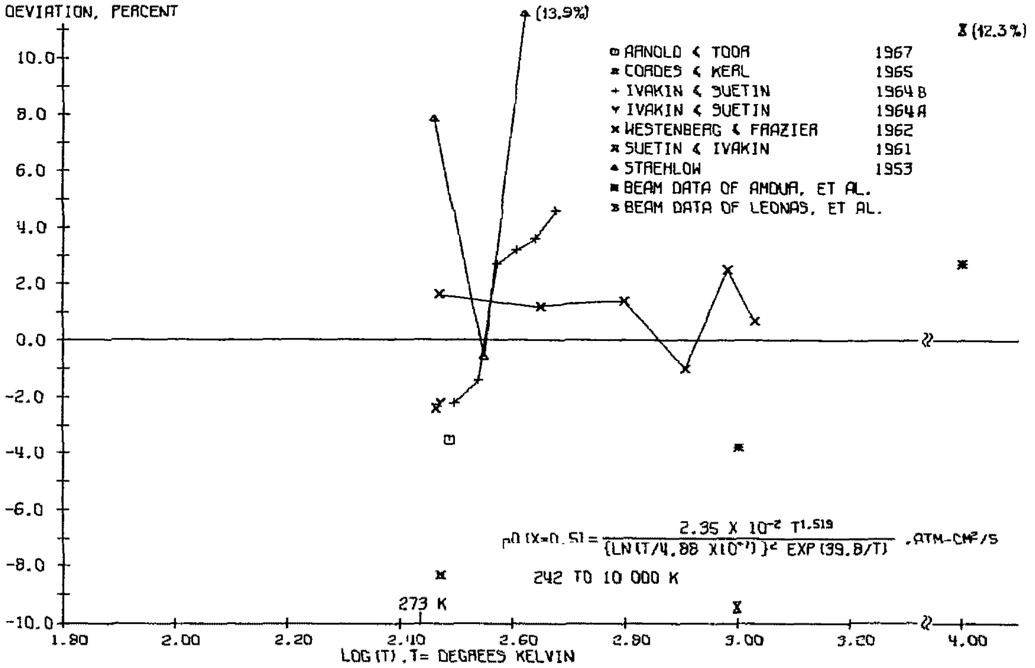

FIGURE 32. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Argon

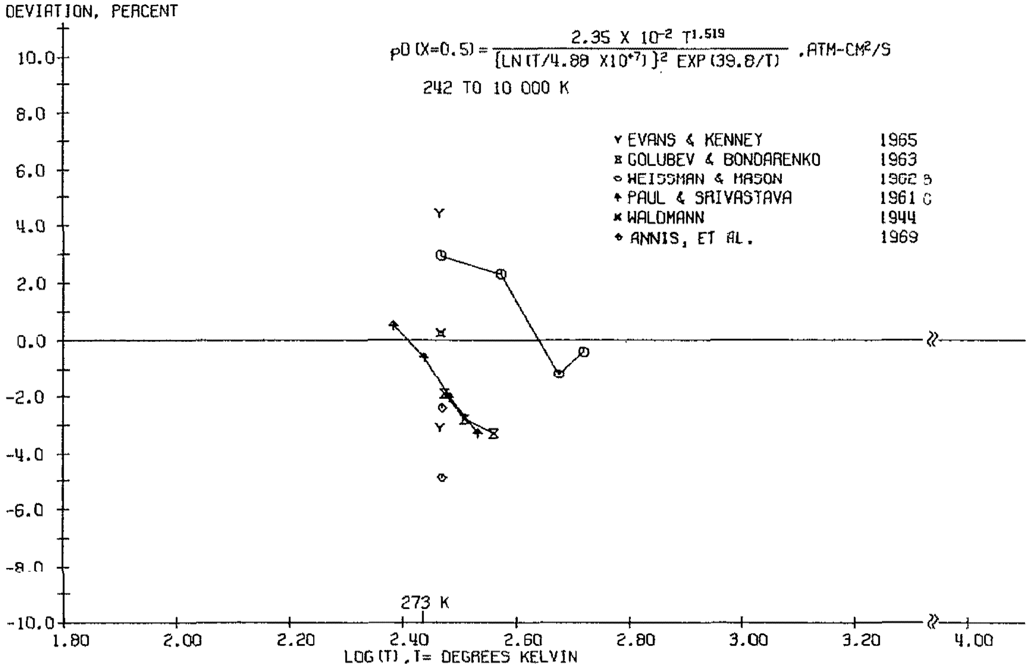

FIGURE 33. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Argon

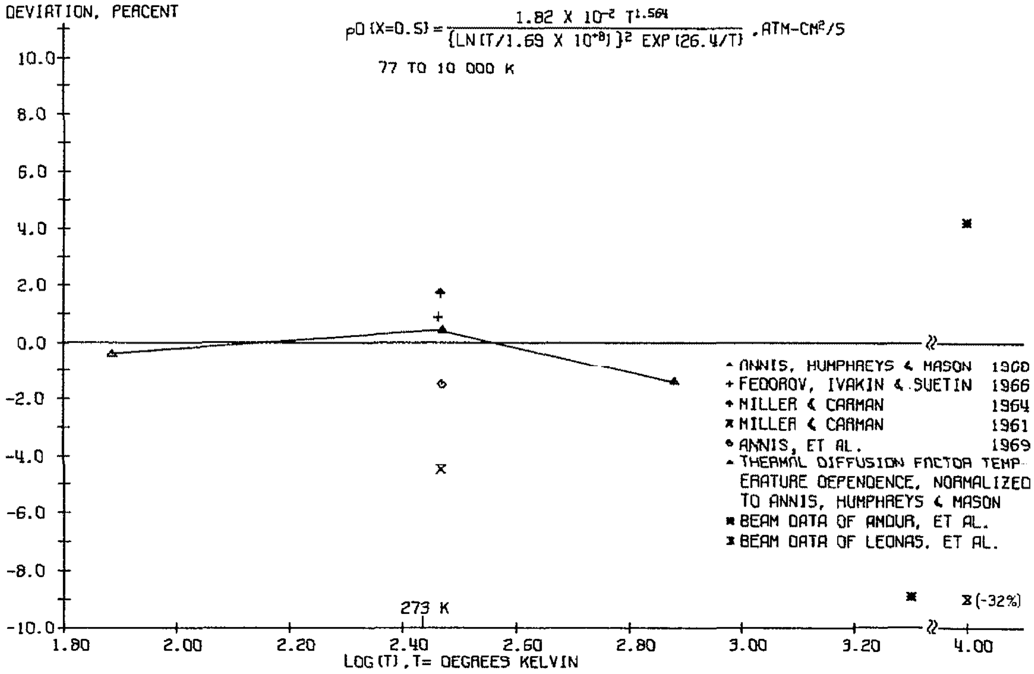

FIGURE 34. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Krypton

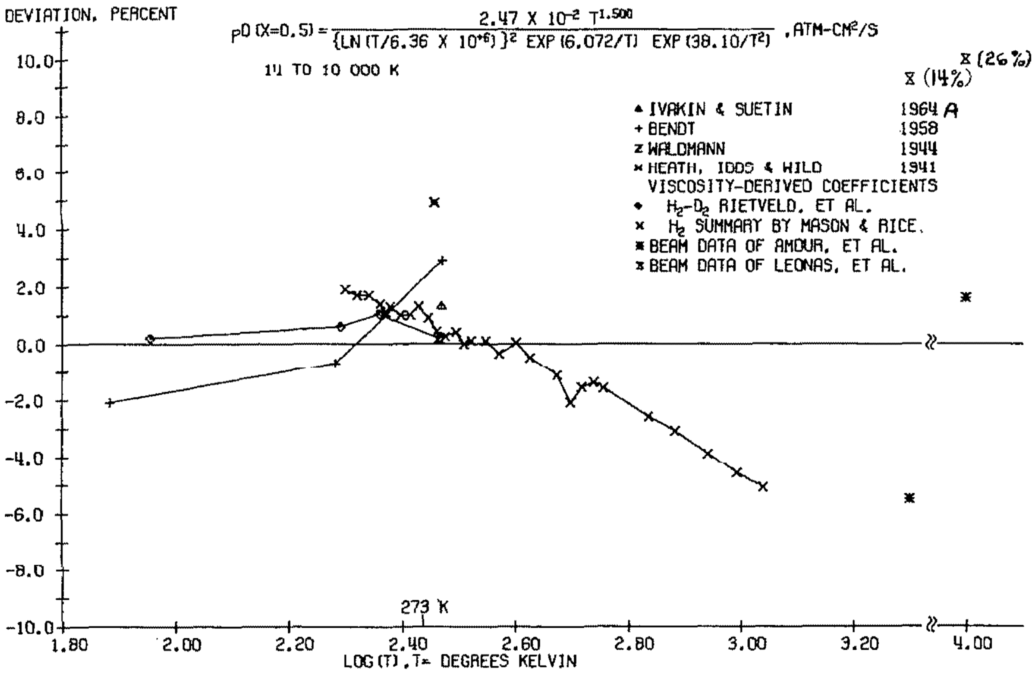

FIGURE 35. *Deviations of diffusion coefficients from reference equation.*

Hydrogen–Deuterium

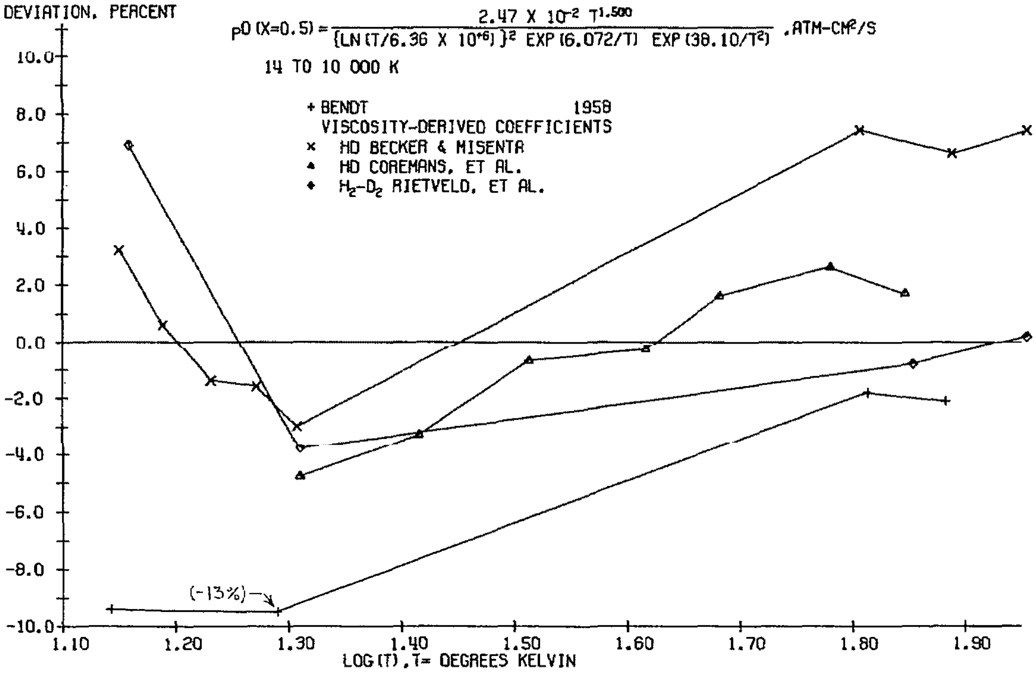

FIGURE 36. *Deviations of diffusion coefficients from reference equation.*

Hydrogen–Deuterium

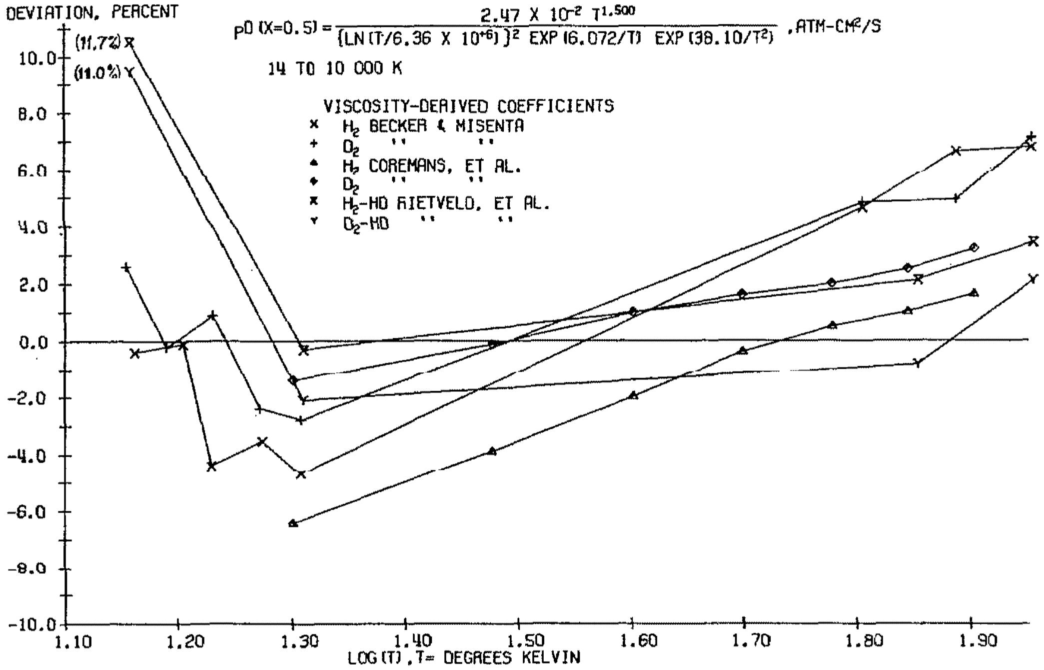

FIGURE 37. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Deuterium

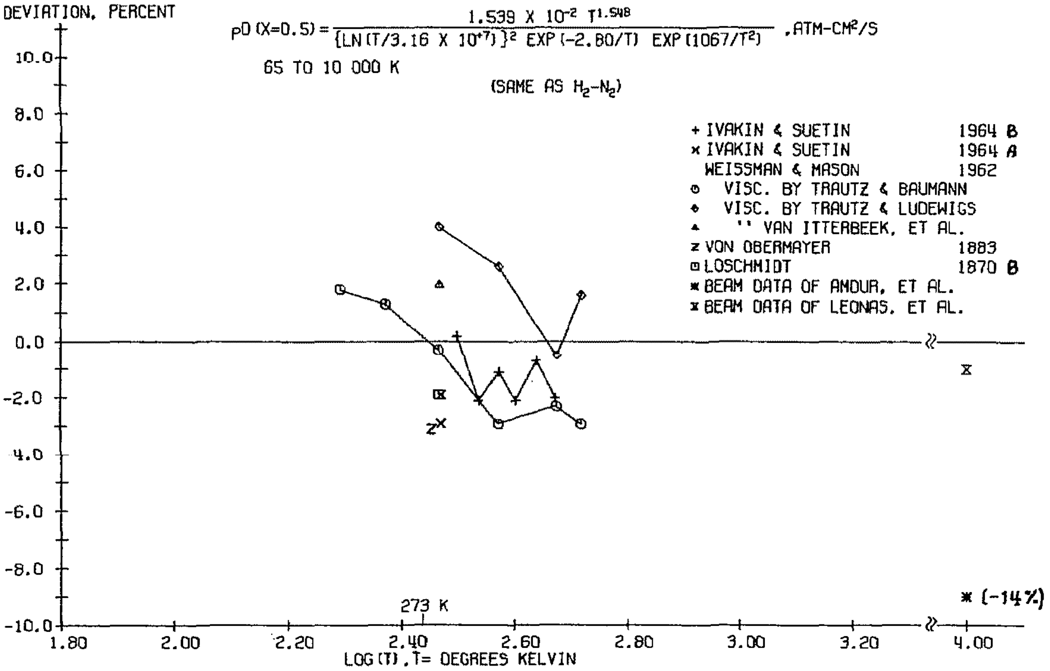

FIGURE 38. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Carbon monoxide

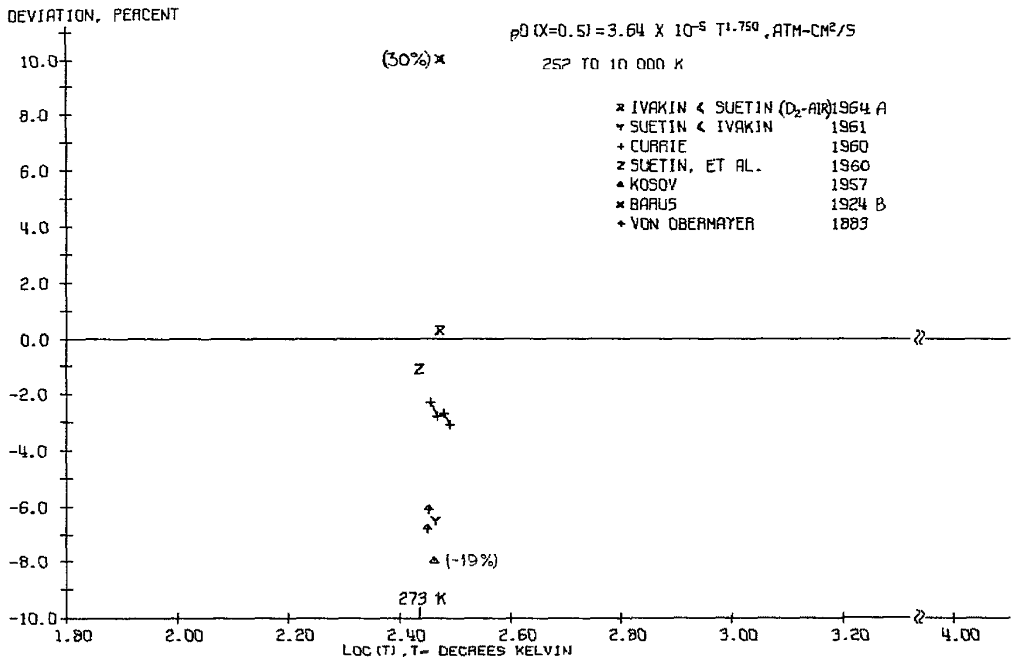

FIGURE 39. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Air

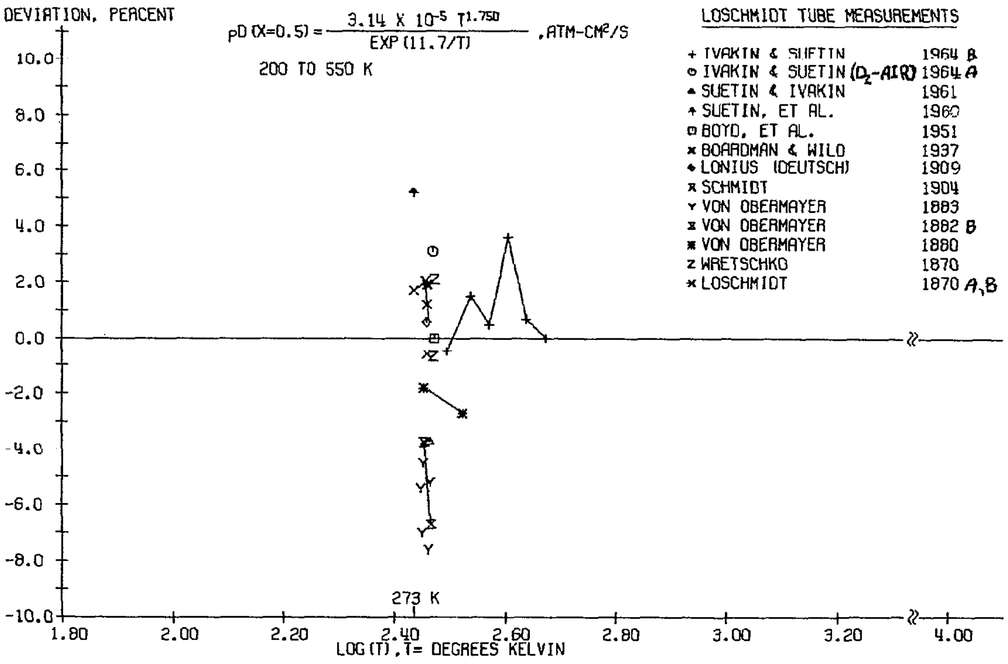

FIGURE 40. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Carbon dioxide

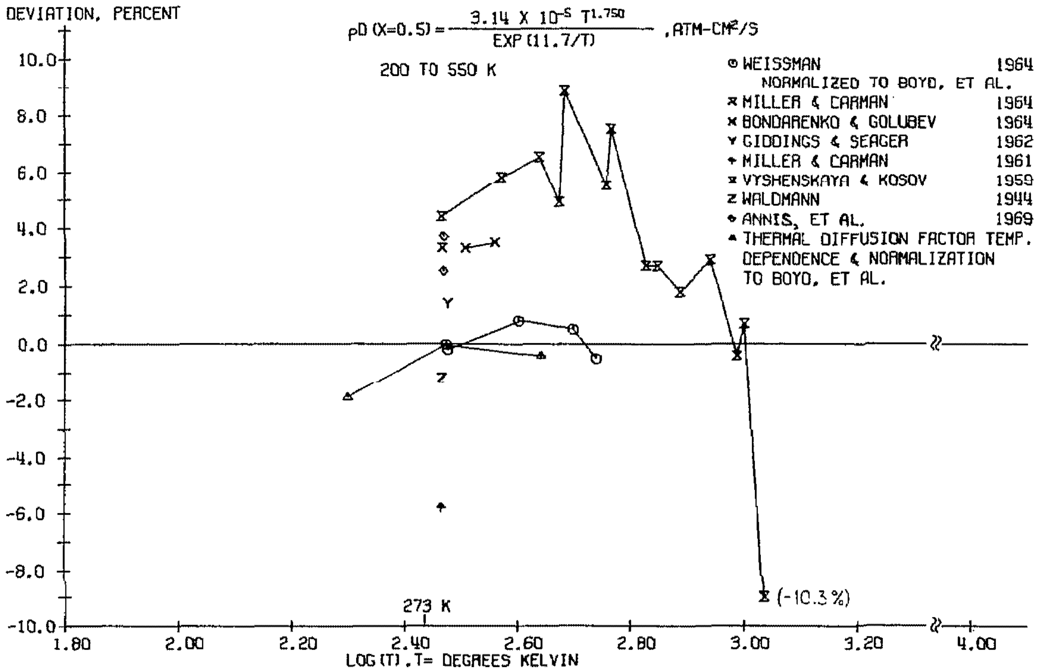

FIGURE 41. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Carbon dioxide

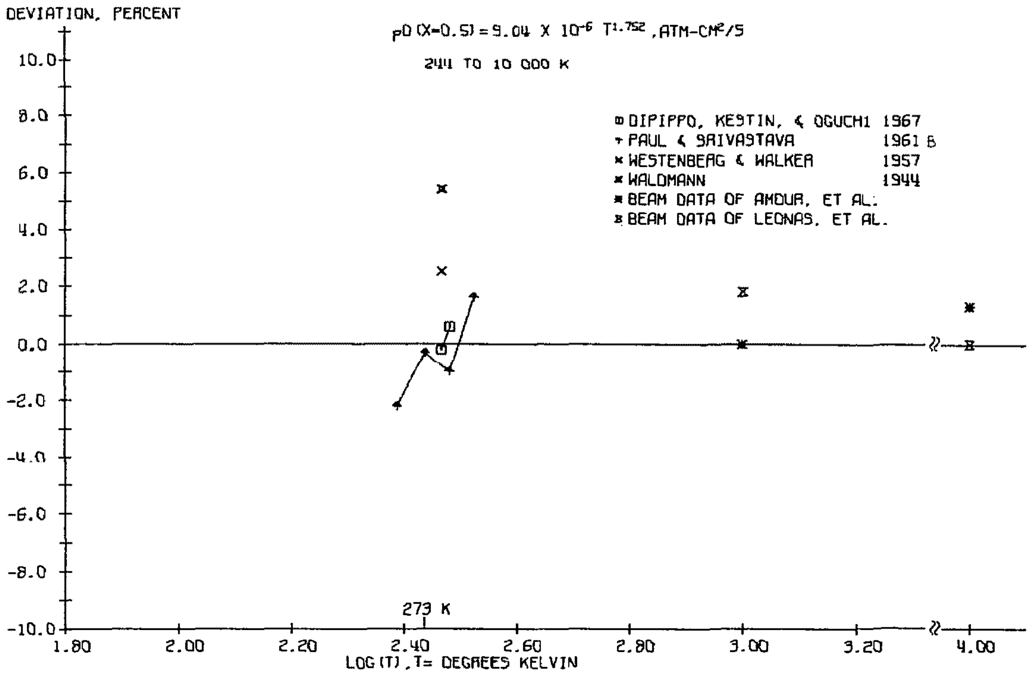

FIGURE 42. *Deviations of diffusion coefficients from reference equation.*

Nitrogen—Argon

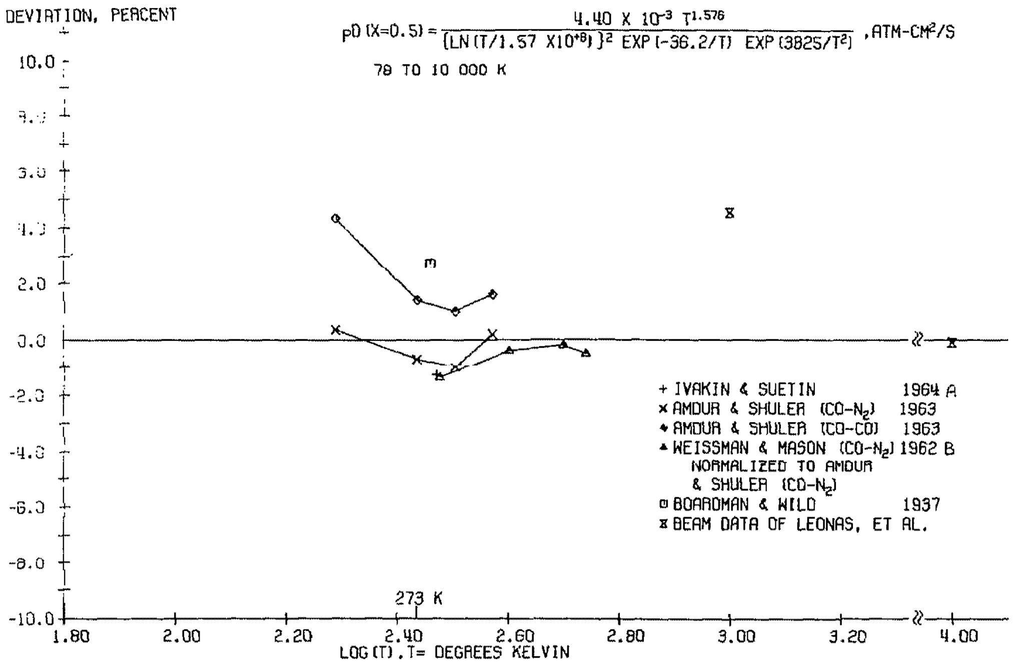

FIGURE 43. *Deviations of diffusion coefficients from reference equation.*

Carbon monoxide—Nitrogen

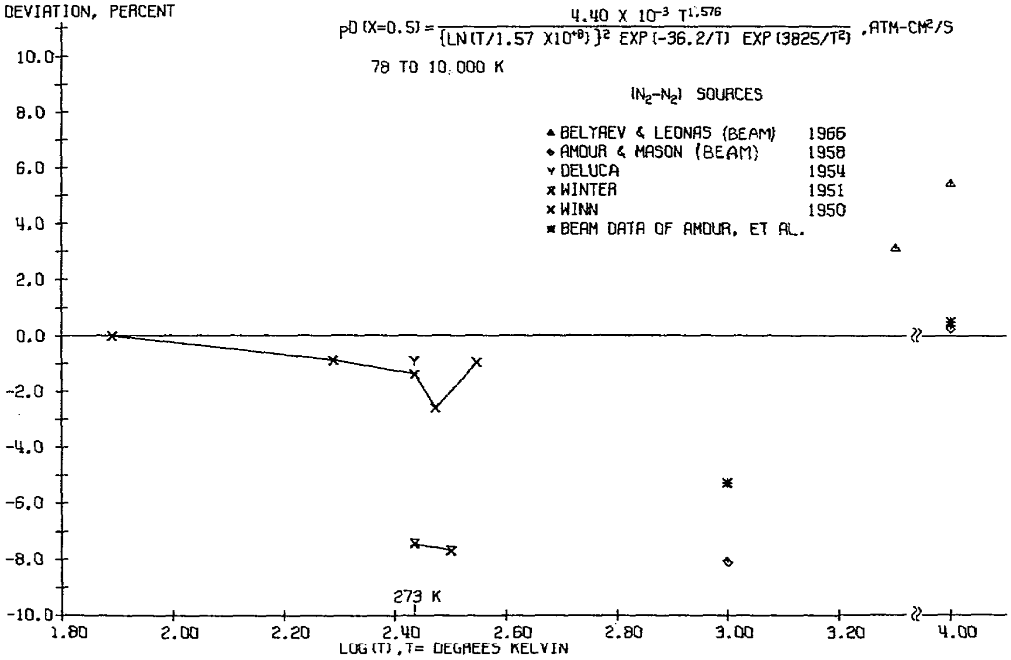

FIGURE 44. *Deviations of diffusion coefficients from reference equation.*

Carbon monoxide—Nitrogen

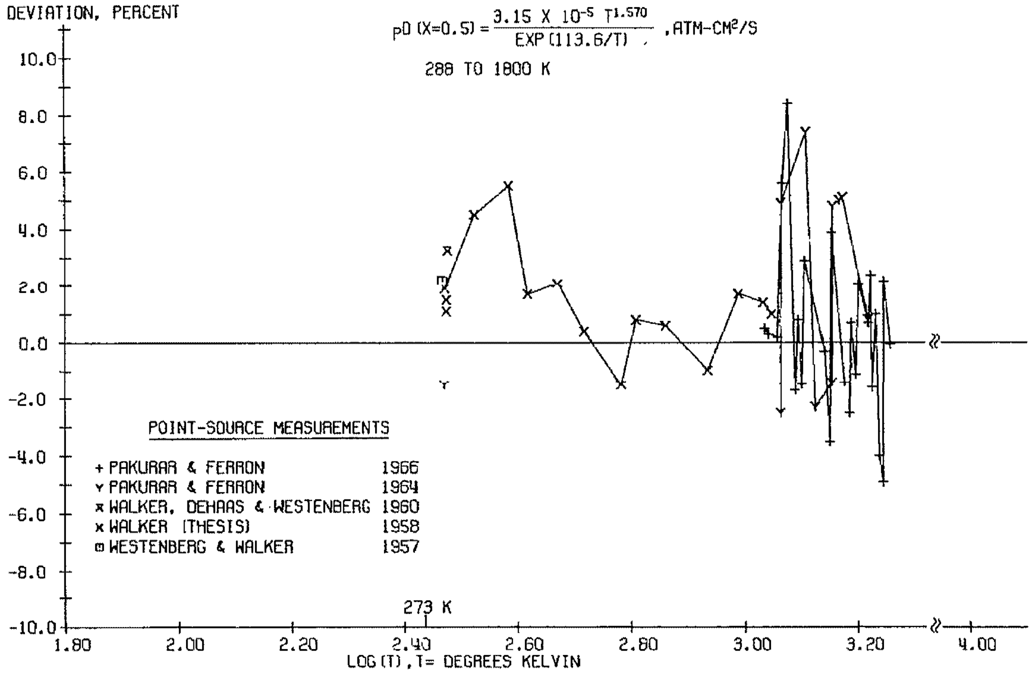

FIGURE 45. *Deviations of diffusion coefficients from reference equation.*

Nitrogen—Carbon dioxide

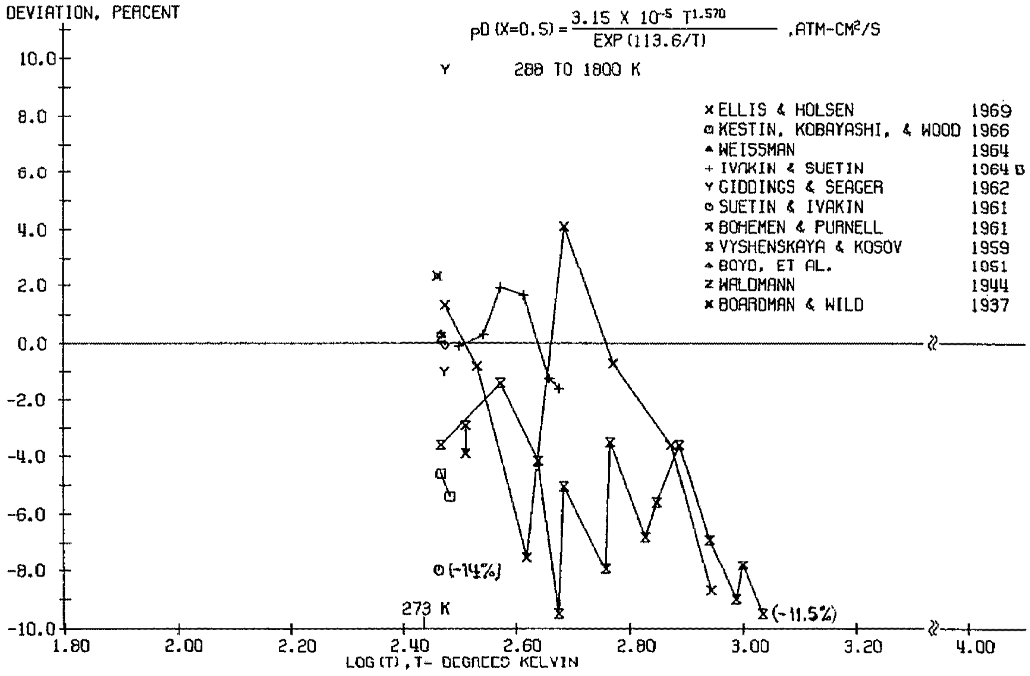

FIGURE 46. *Deviations of diffusion coefficients from reference equation.*

Nitrogen—Carbon dioxide

lack of measurements at low temperatures and asymmetry between the CO and N_2 molecules which may lead to small differences between the potentials.

He-O_2. This gas pair is a borderline system of Group II. He-O_2 was assigned to Group II on the strength of the two-bulb measurements by Paul and Srivastava (1961 a) and the consistency of its $\mathscr{D}_{12}$ with those of He$-N_2$ and He$-$air.

He-air and H_2-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-N_2 and He-O_2, and H_2-air from H_2-N_2 and H_2-O_2. The reliable determinations of $\mathscr{D}_{12}$ have been by the closed-tube method; namely, for He-air the room temperature point from Fedorov et al. (1966) and for H_2-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 H_2-air system was not assigned to Group I along with H_2-N_2, but to Group II.

Each lower temperature limit for the reference equations of He-air and H_2-air has been set by data of He-O_2 (244 K) and H_2-O_2 (252 K), respectively. But the lower temperature limit for He-N_2 is 77 K, and for H_2-N_2 it is 65 K. In order to estimate $\mathscr{D}_{12}$ for He-air at lower temperatures, if necessary, it is suggested that the ratio of $\mathscr{D}_{12}$ for He-O_2 to He-N_2 at room temperature be taken as a constant, independent of temperature, and a similar procedure used for H_2-air. By applying this ratio the correlation range can be extended for He-O_2 and H_2-O_2, as well as He-air and H_2-air to the lower temperature limits for He-N_2 (77 K) and for H_2-N_2 (65 K).

He-CO_2. 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-CO_2 as a Group II gas pair.

H_2-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).

H_2-Ne. This gas pair is a borderline Group II system because only one set of direct measurements of $\mathscr{D}_{12}$ is available, obtained by the two-bulb method (Paul and Srivastava, 1961 c).

H_2-Ar. There are many independent determinations of $\mathscr{D}_{12}$ for H_2-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 $\mathscr{D}_{12}$. The only direct molecular-beam measurement for H_2-Ar gives a potential which is suspected to be too great (Colgate et al., 1969). Thus the molecular-beam potential for H_2-Ar was obtained by application of the combination rules, see table 21.

H_2-Kr. The gas pair H_2-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 H_2-Kr is considered a borderline system.

H_2-D_2. Most of the $\mathscr{D}_{12}$ for H_2-D_2 have been calculated from viscosity measurements of the H_2 isotopes or mixtures thereof, see figures 36 and 37. At low temperatures the viscosity-derived $\mathscr{D}_{12}$ are considered more reliable than the direct measurements. None of the direct measurements of $\mathscr{D}_{12}$ 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 $s=1.500$.

H_2-CO_2. 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 $\mathscr{D}_{12}$ reported by Weissman (1964) were normalized to the datum of Boyd et al. (1951) because the points seemed systematically high by about 5 percent.

N_2-Ar. The N_2-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.

N_2-CO. On the basis of two-bulb measurements by Winn (1950) and closed-tube measurements by Amdur and Shuler (1963), the gas pair N_2-CO was placed in Group II. Since N_2 and CO are isosteric molecules, the N_2-N_2, CO-CO, and N_2-CO data could all be used for $\mathscr{D}_{12}$. Determinations of $\mathscr{D}_{12}$ from molecular-beam measurements have been published by Belyaev and Leonas (1966) and Amdur and Mason (1958) for N_2-N_2, and are in excellent agreement with the present calculations, see figure 44. The viscosity-derived $\mathscr{D}_{12}$ 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).

N_2-CO_2. 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 $\mathscr{D}_{12}$ at high temperatures; a similar trend was evident for H_2-CO_2.

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

Weights and Potentials. The equimolar values of $\mathscr{D}_{12}$ 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 CO_2, no potentials are listed for the systems air-(Ar, CH_4, CO, SF_6) and CO_2-(Ar, CO, O_2, air, N_2O, SF_6). 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 CH_4 and SF_6, but both of these gases have been used in experiments 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 H_2-CH_4; but the CH_4-Ar potential was used for the heavier gas pairs with methane; CH_4-(N_2,O_2,SF_6). If the He-CH_4 potential had been used for CH_4-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 H_2-O_2, CH_4-O_2, CO-Kr, CO-O_2, and N_2-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.

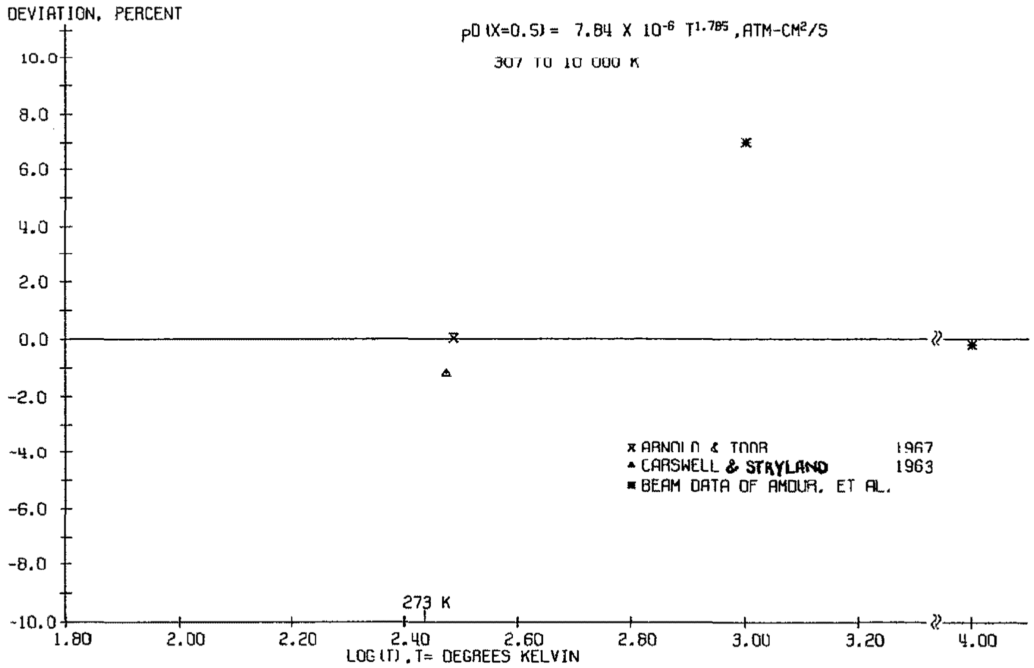

FIGURE 47. *Deviations of diffusion coefficients from reference equation.*

Methane—Argon

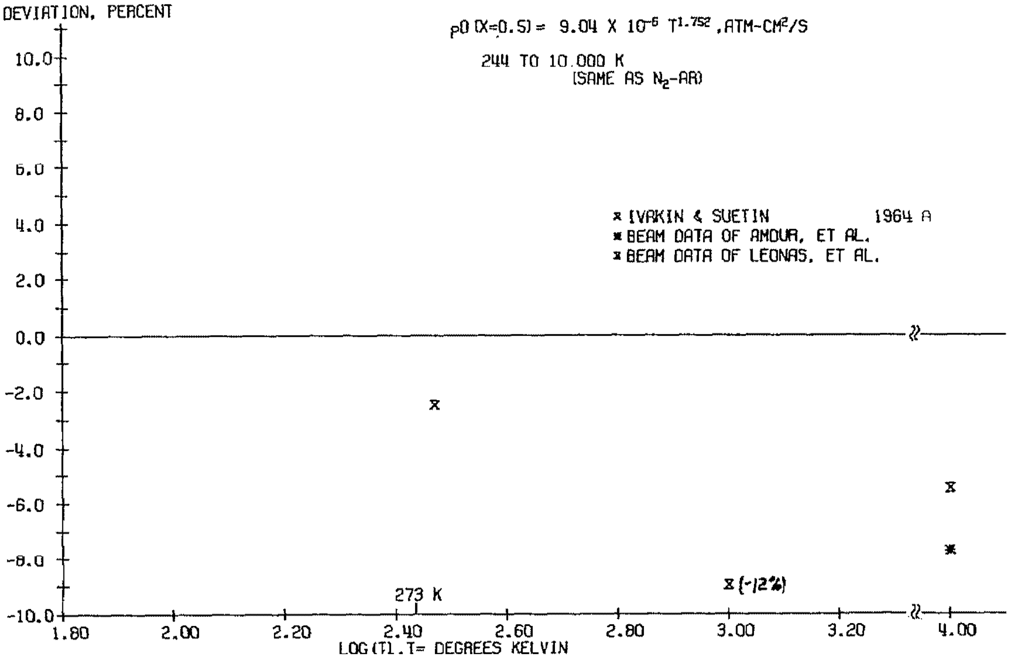

FIGURE 48. *Deviations of diffusion coefficients from reference equation.*

Carbon monoxide—Argon

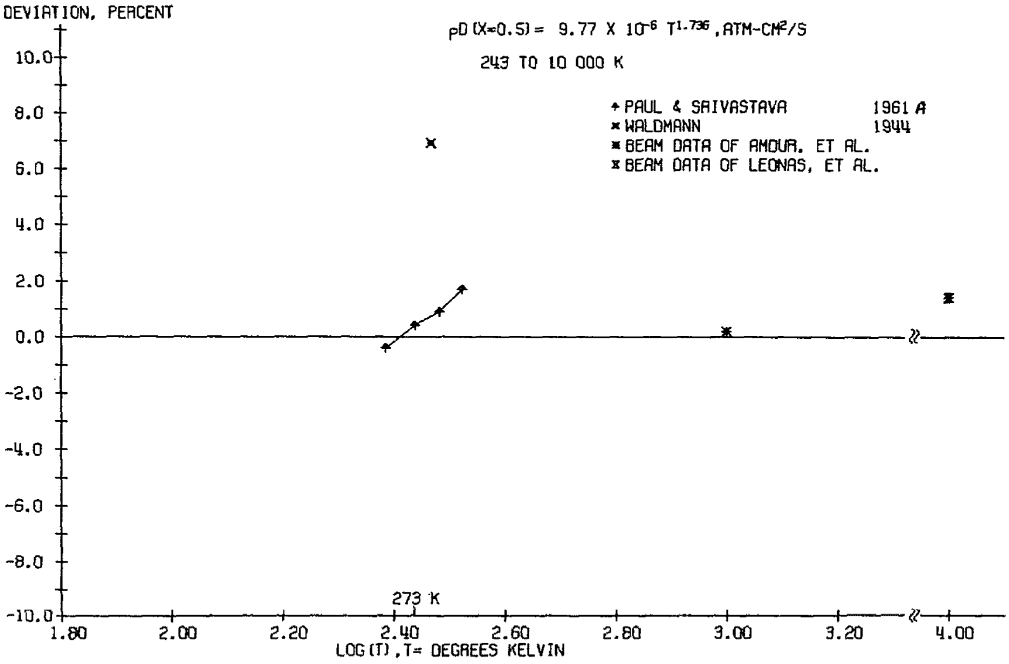

FIGURE 49. *Deviations of diffusion coefficients from reference equation.*

Oxygen—Argon

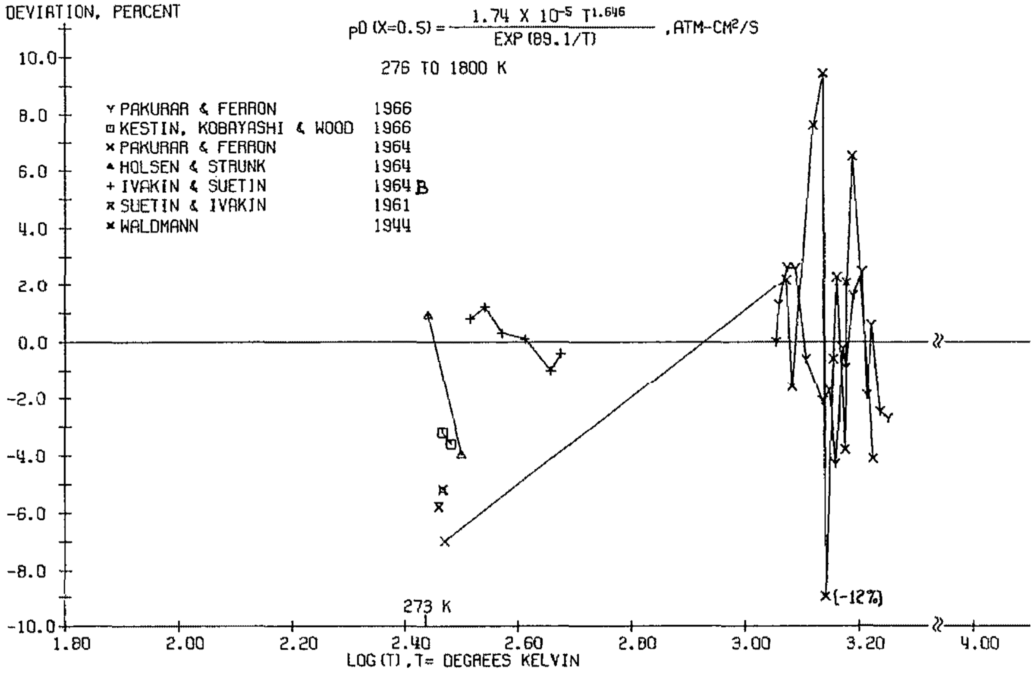

FIGURE 50. *Deviations of diffusion coefficients from reference equation.*

Argon—Carbon dioxide

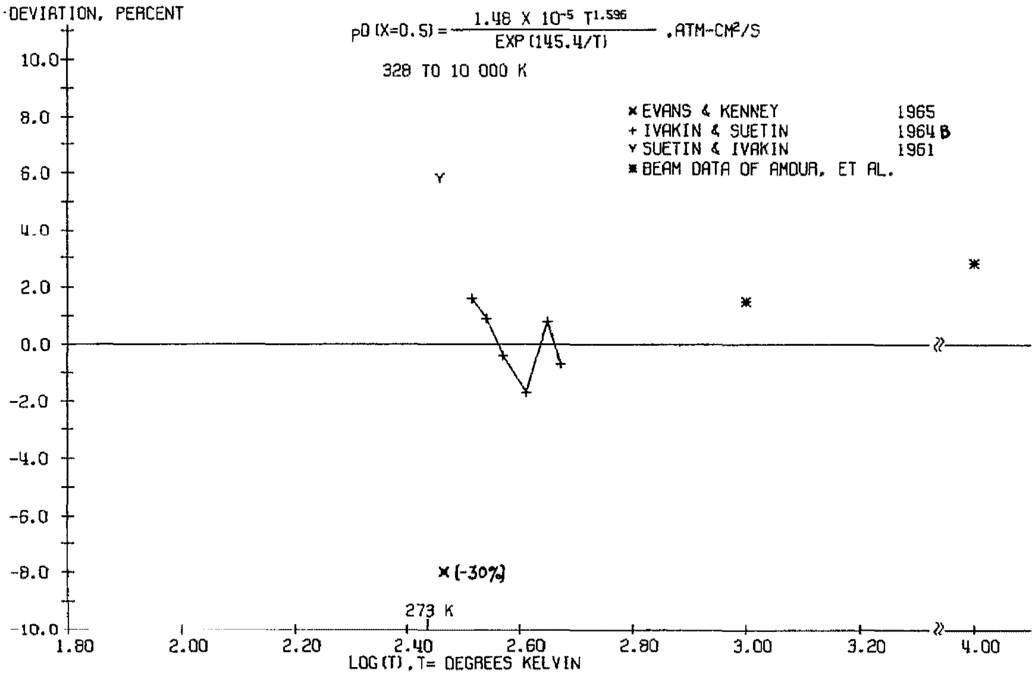

FIGURE 51. *Deviations of diffusion coefficients from reference equation.*

Argon—SF_6

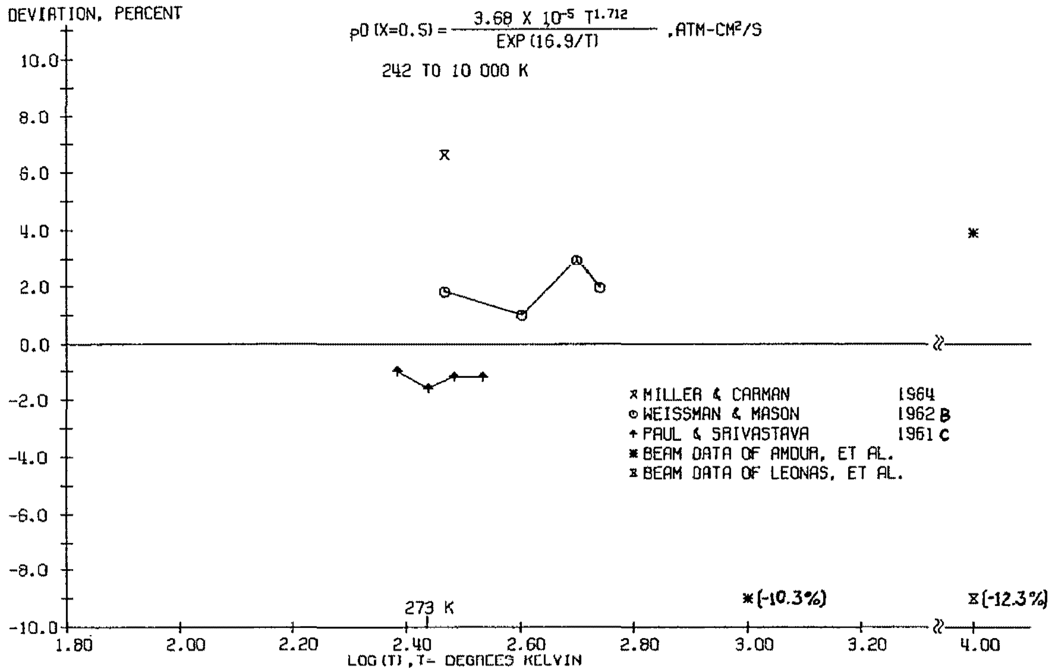

FIGURE 52. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Xenon

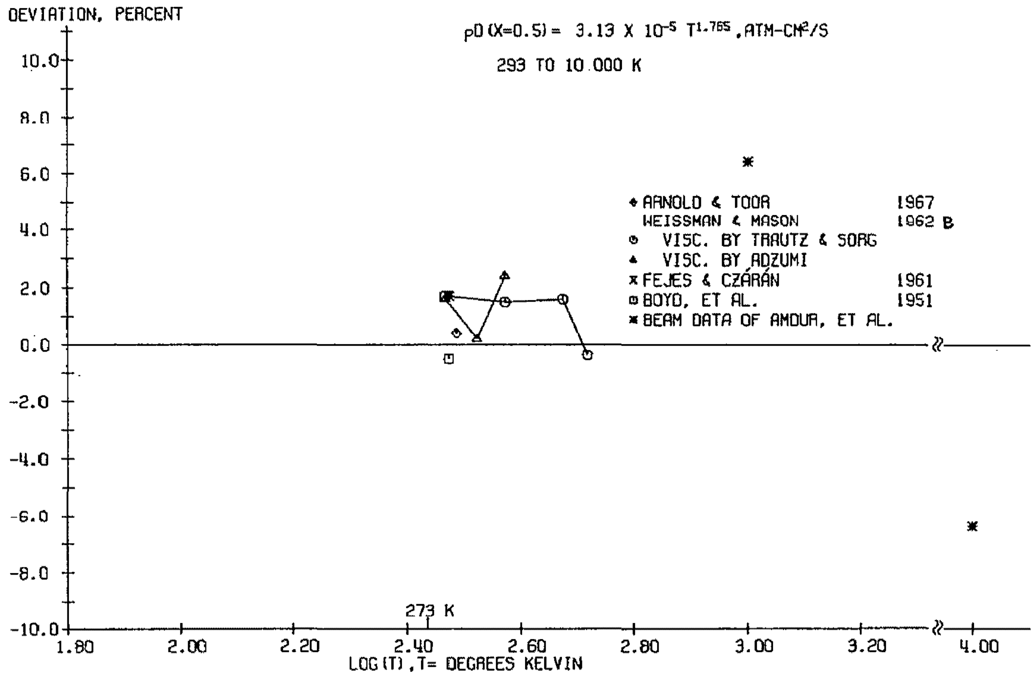

FIGURE 53. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Methane

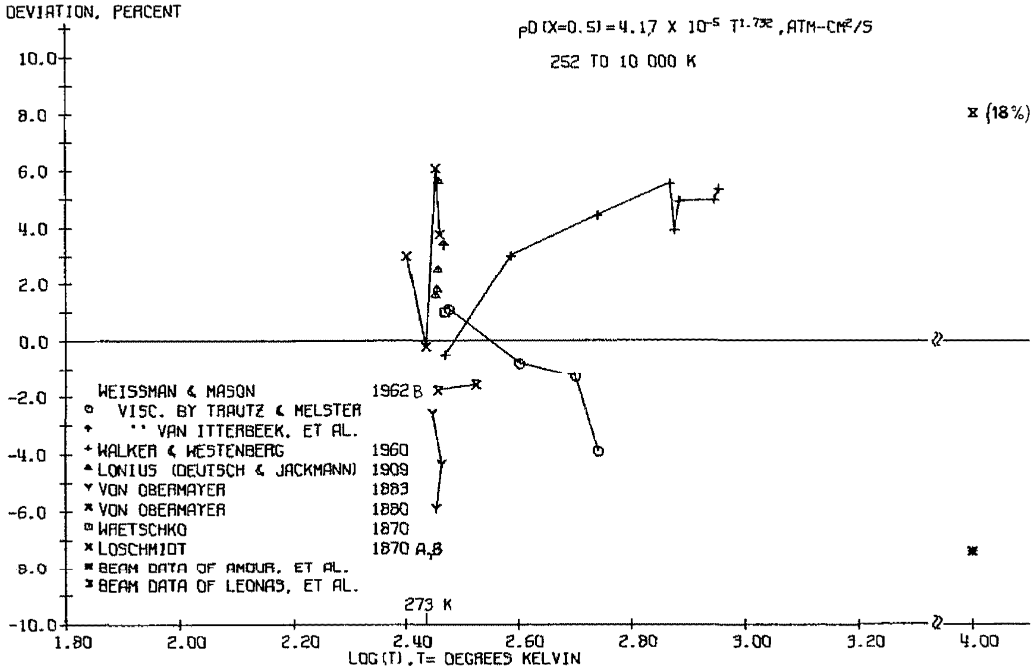

FIGURE 54. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—Oxygen

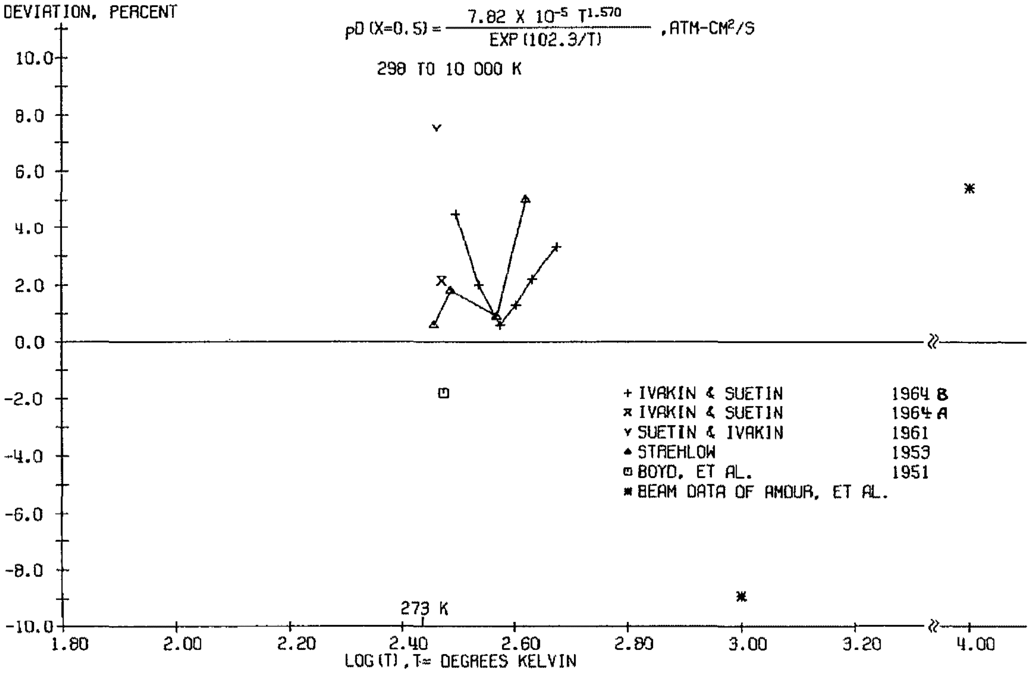

FIGURE 55. *Deviations of diffusion coefficients from reference equation.*

Hydrogen—SF_6

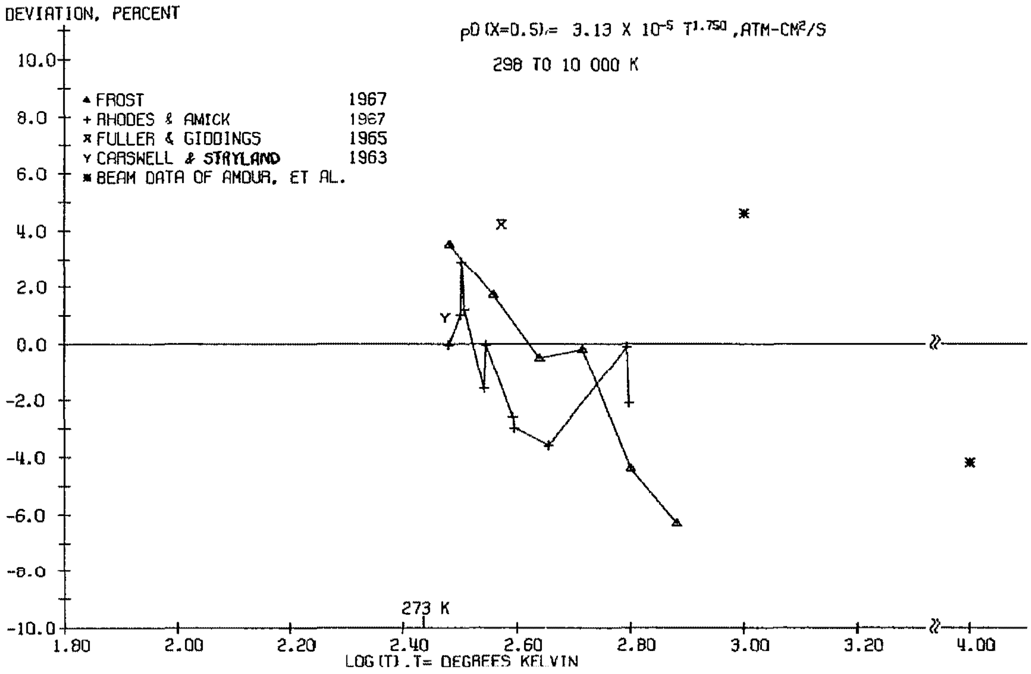

FIGURE 56. *Deviations of diffusion coefficients from reference equation.*

Helium—Methane

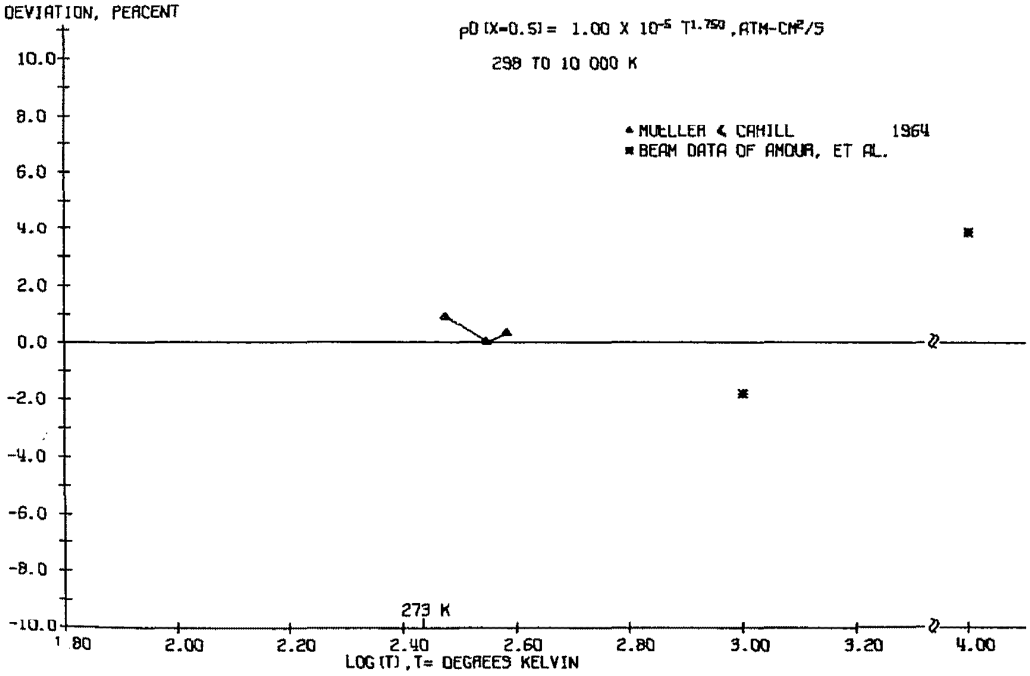

FIGURE 57. *Deviations of diffusion coefficients from reference equation.*

Methane—Nitrogen

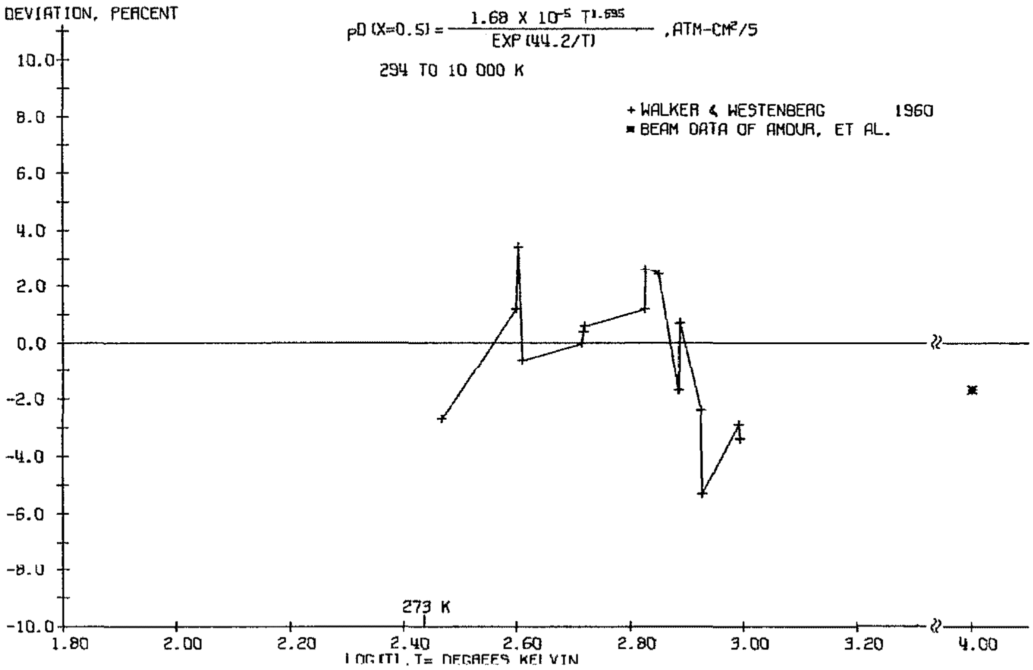

FIGURE 58. *Deviations of diffusion coefficients from reference equation.*

Methane—Oxygen

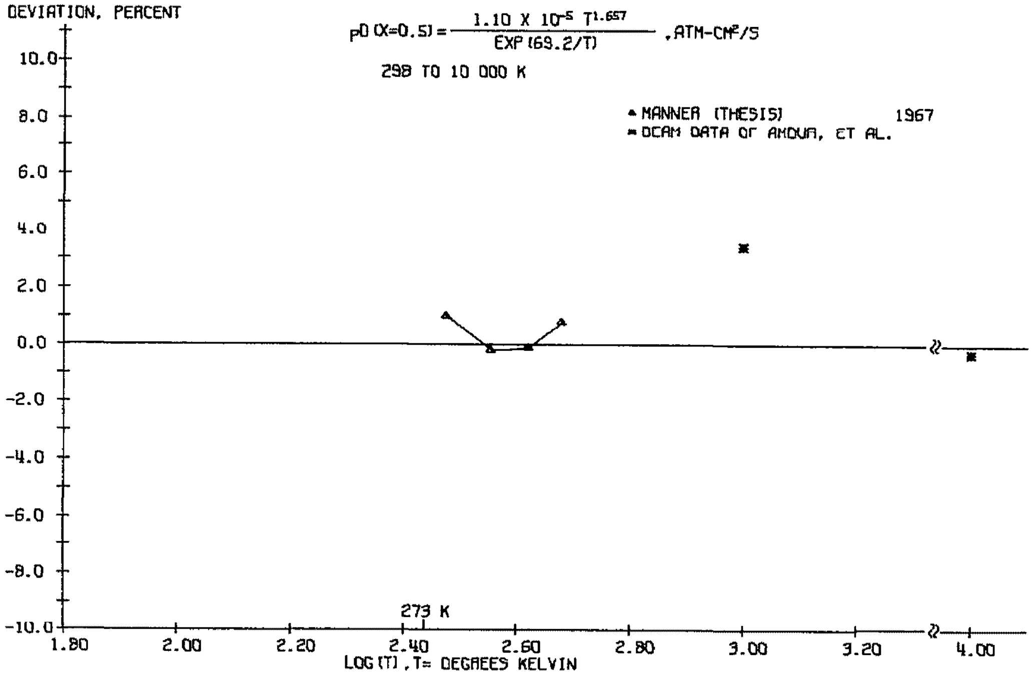

FIGURE 59. *Deviations of diffusion coefficients from reference equation.*

Methane—SF_6

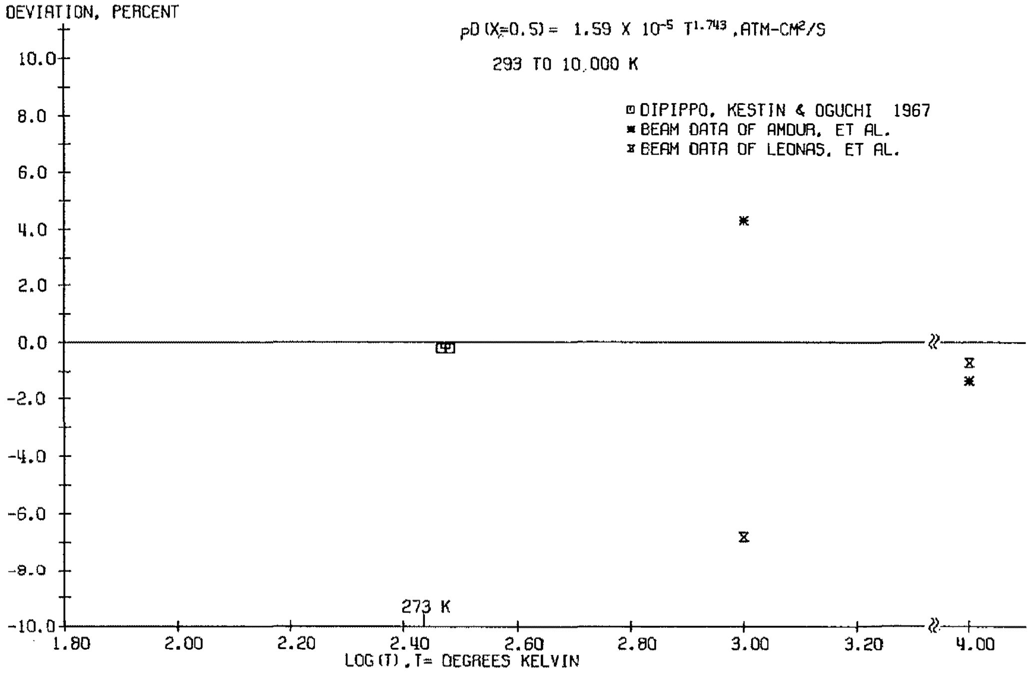

FIGURE 60. *Deviations of diffusion coefficients from reference equation.*

Neon—Nitrogen

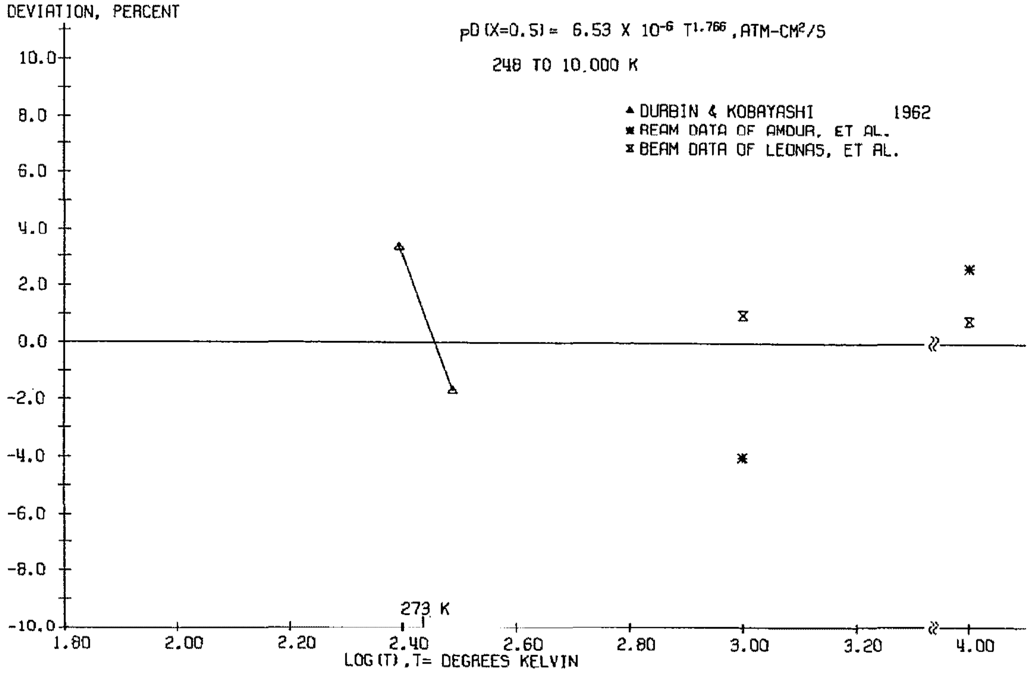

FIGURE 61. *Deviations of diffusion coefficients from reference equation.*

Nitrogen—Krypton

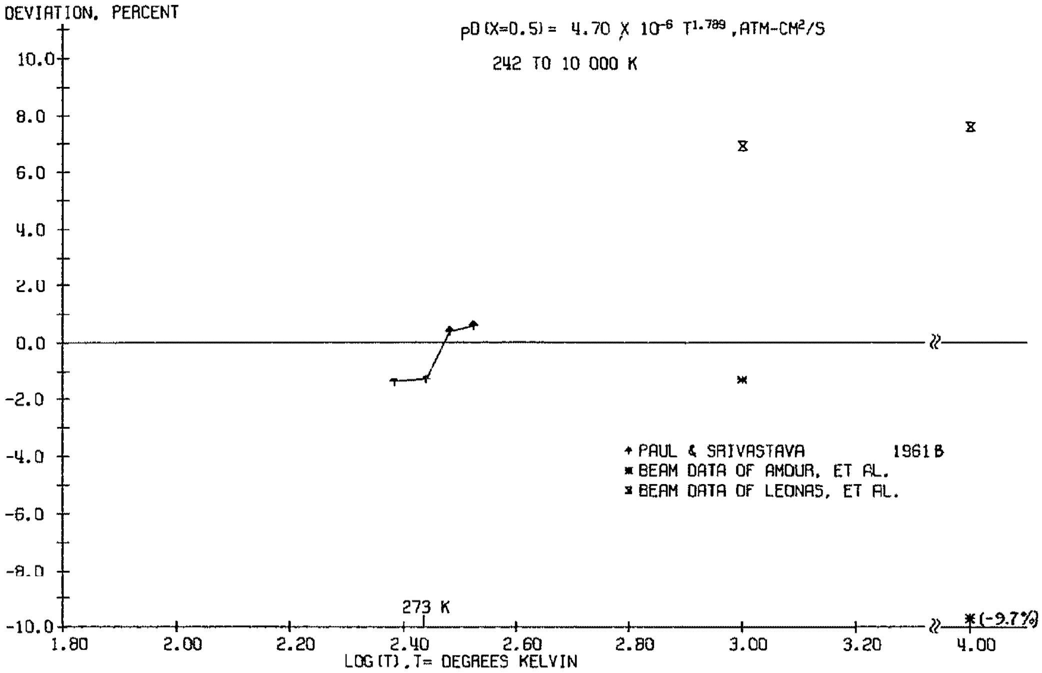

FIGURE 62. *Deviations of diffusion coefficients from reference equation.*

Nitrogen—Xenon

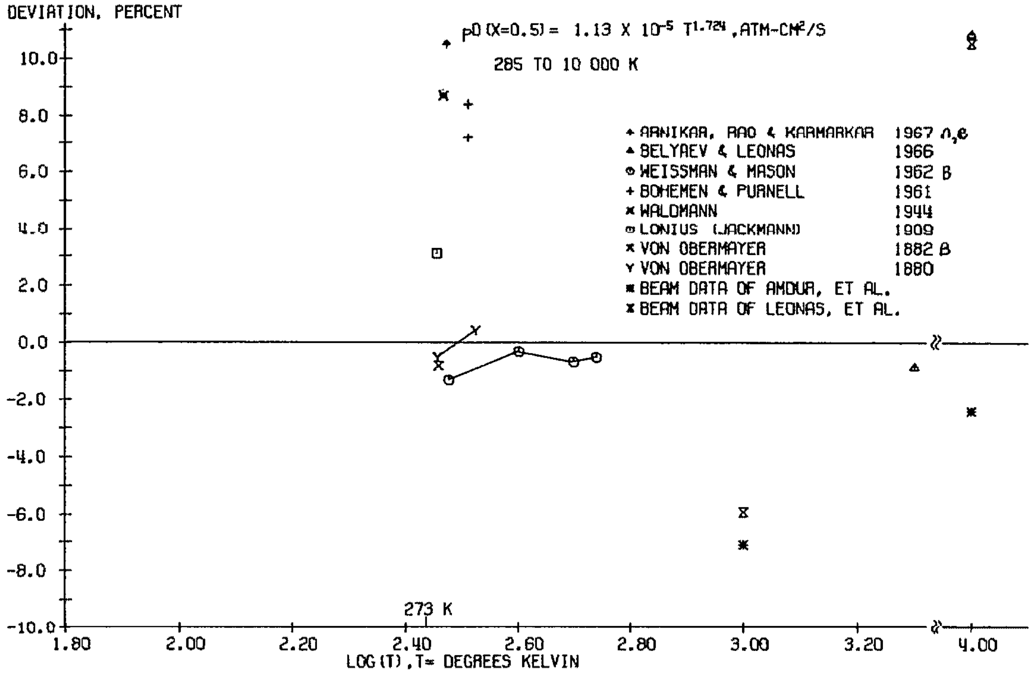

FIGURE 63. *Deviations of diffusion coefficients from reference equation.*

Nitrogen—Oxygen

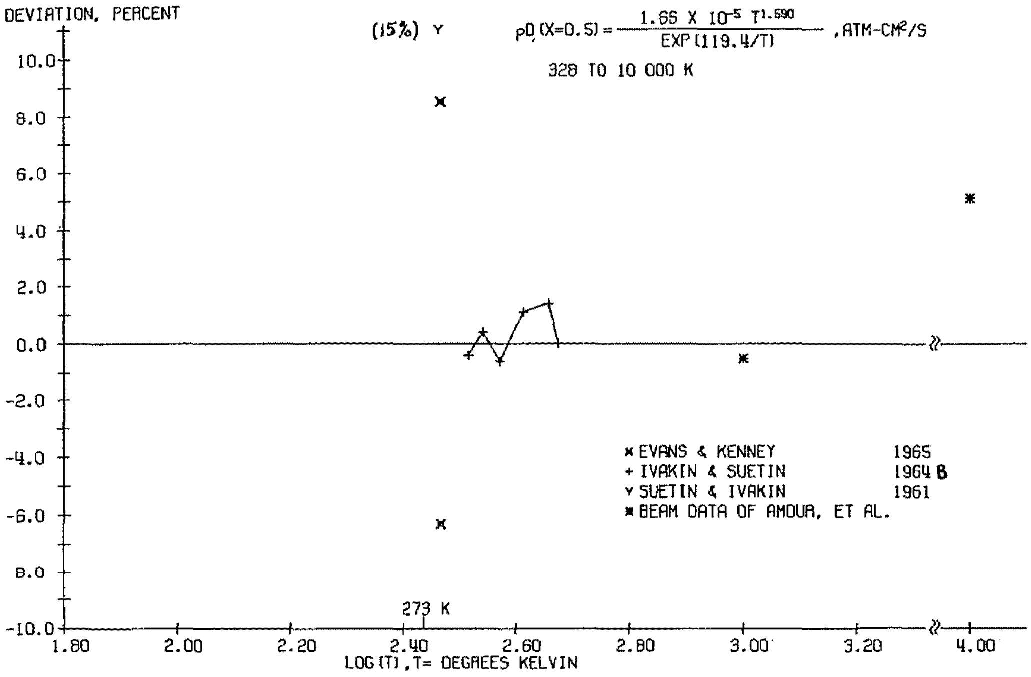

FIGURE 64. *Deviations of diffusion coefficients from reference equation.*

Nitrogen—SF_6

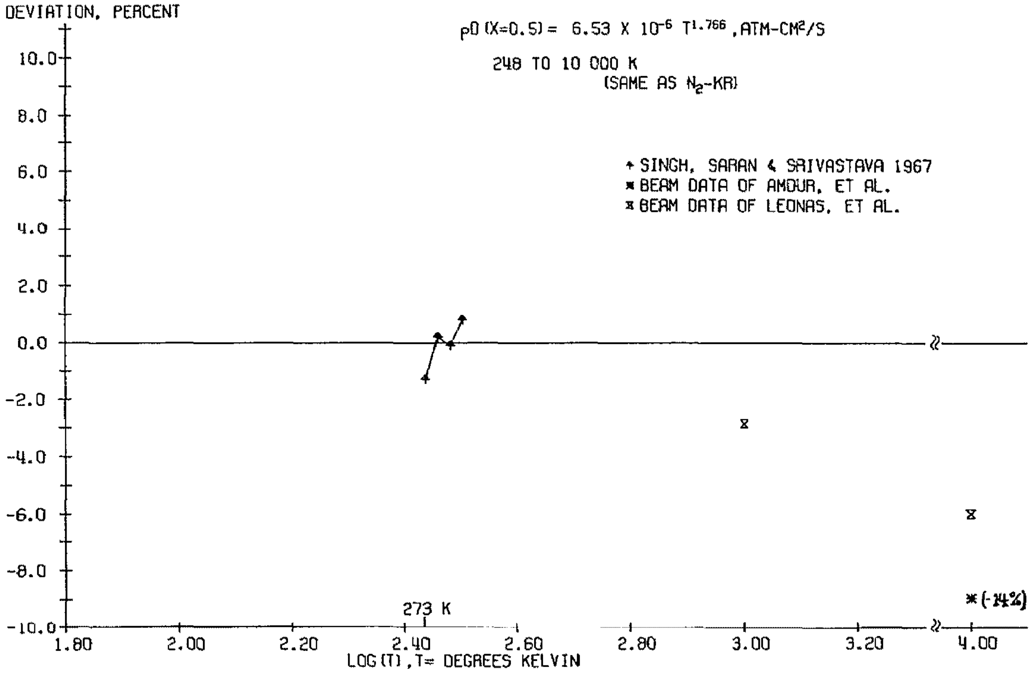

FIGURE 65. *Deviations of diffusion coefficients from reference equation.*

Carbon monoxide—Krypton

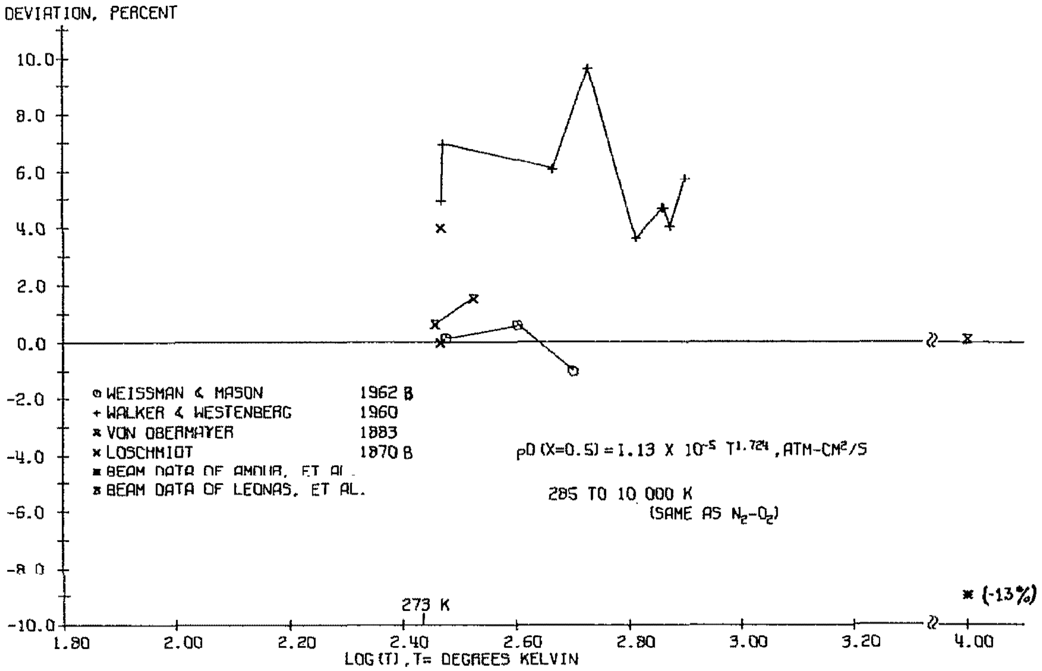

FIGURE 66. *Deviations of diffusion coefficients from reference equation.*

Carbon monoxide—Oxygen

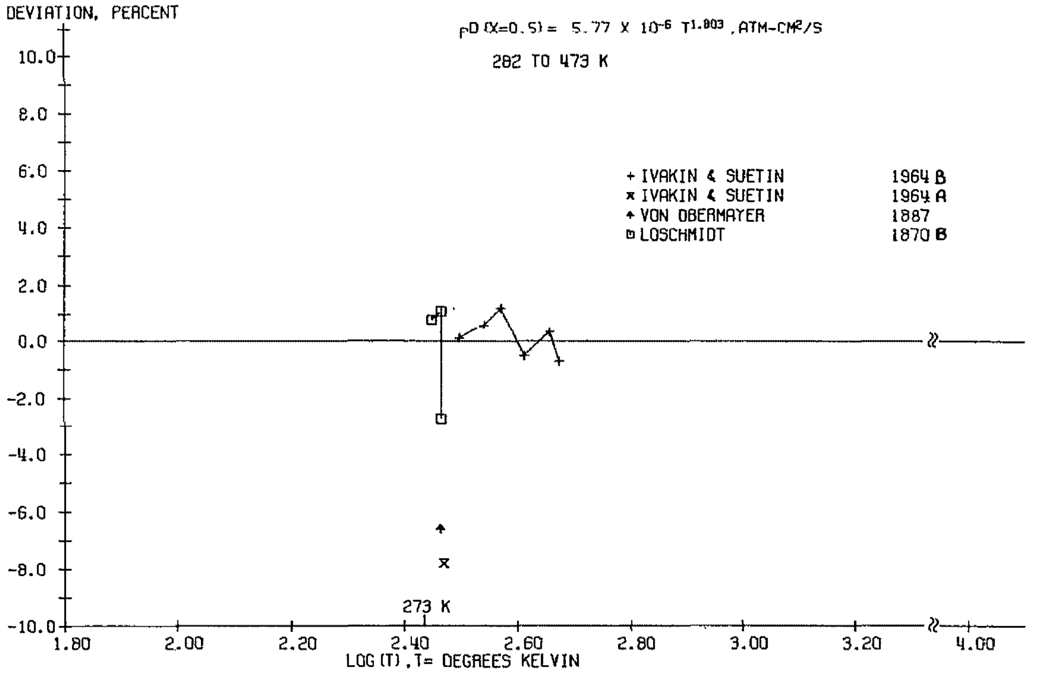

FIGURE 67. *Deviations of diffusion coefficients from reference equation.*

Carbon monoxide—Carbon dioxide

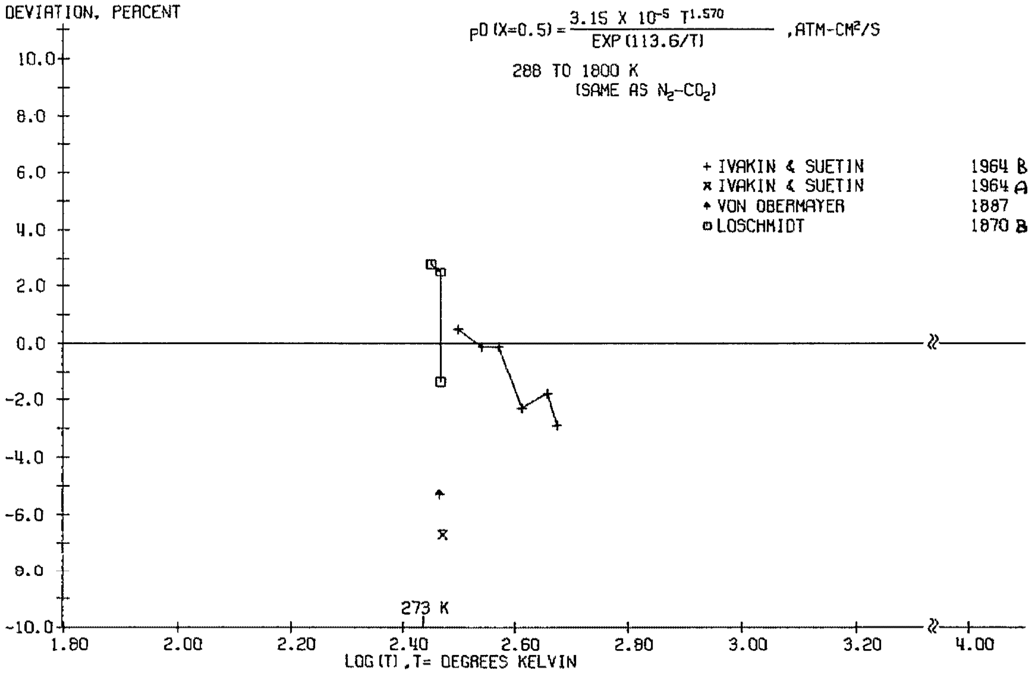

FIGURE 68. *Deviations of diffusion coefficients from reference equation.*

Carbon monoxide—Carbon dioxide

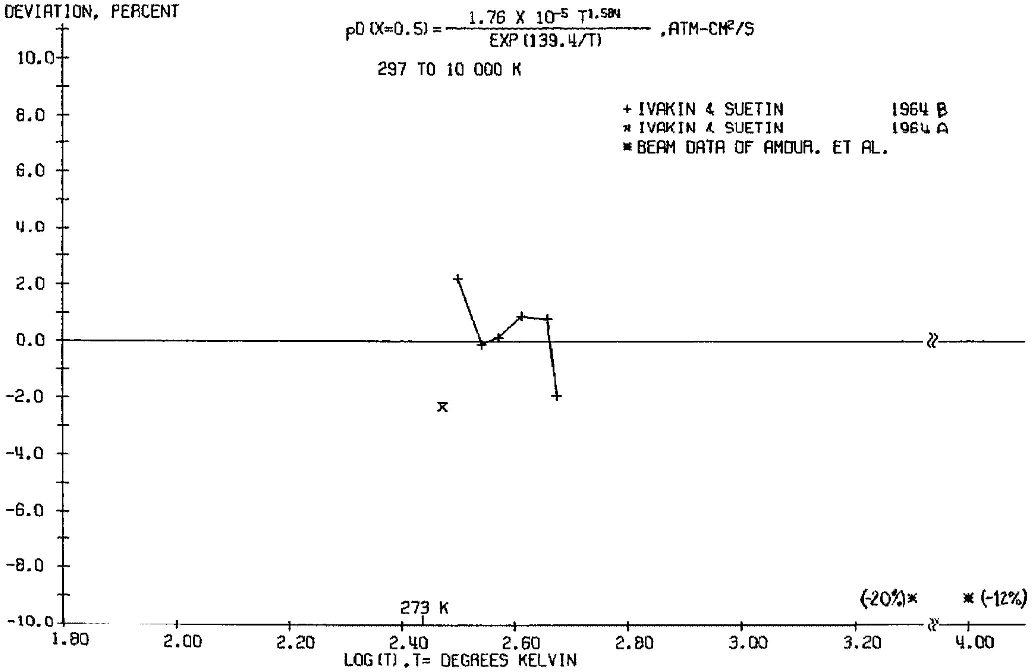

FIGURE 69. *Deviations of diffusion coefficients from reference equation.*

Carbon monoxide—SF_6

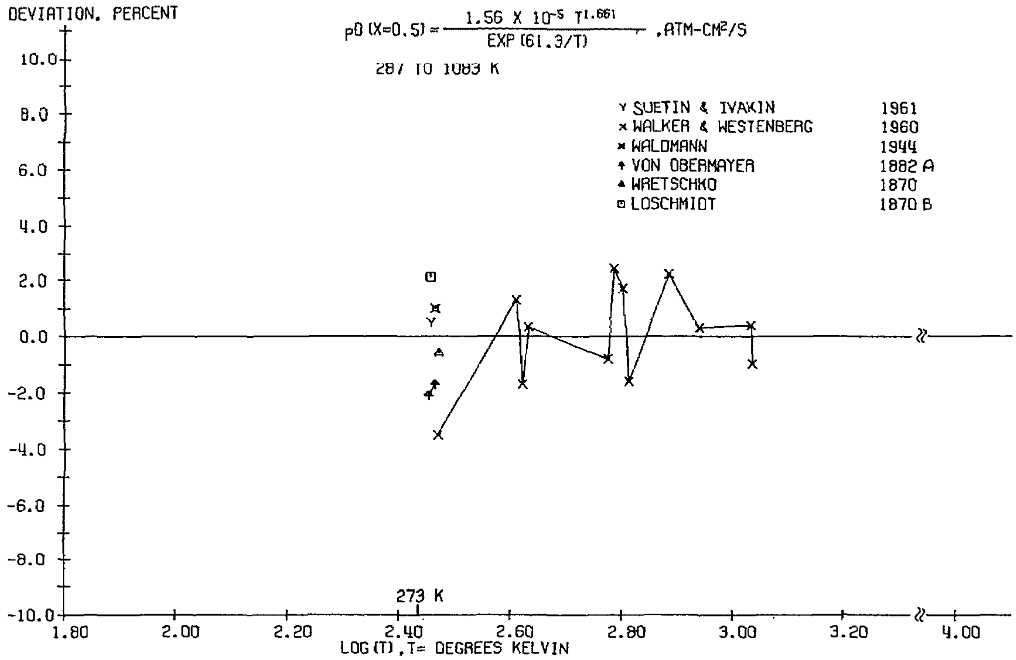

Figure 70. *Deviations of diffusion coefficients from reference equation.*

Oxygen—Carbon dioxide

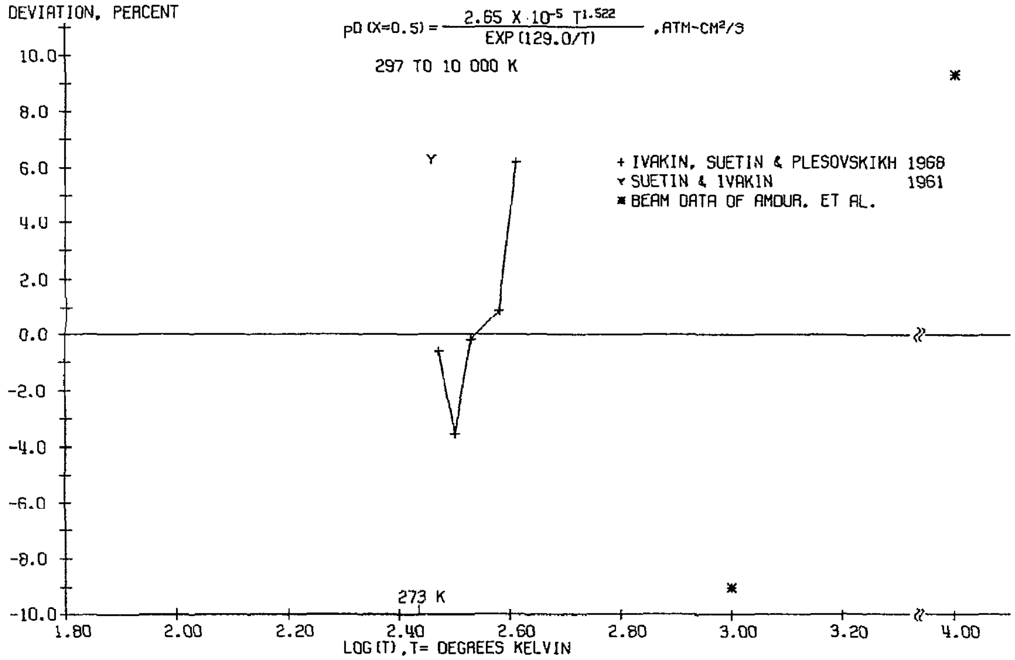

Figure 71. *Deviations of diffusion coefficients from reference equation.*

Oxygen—SF_6

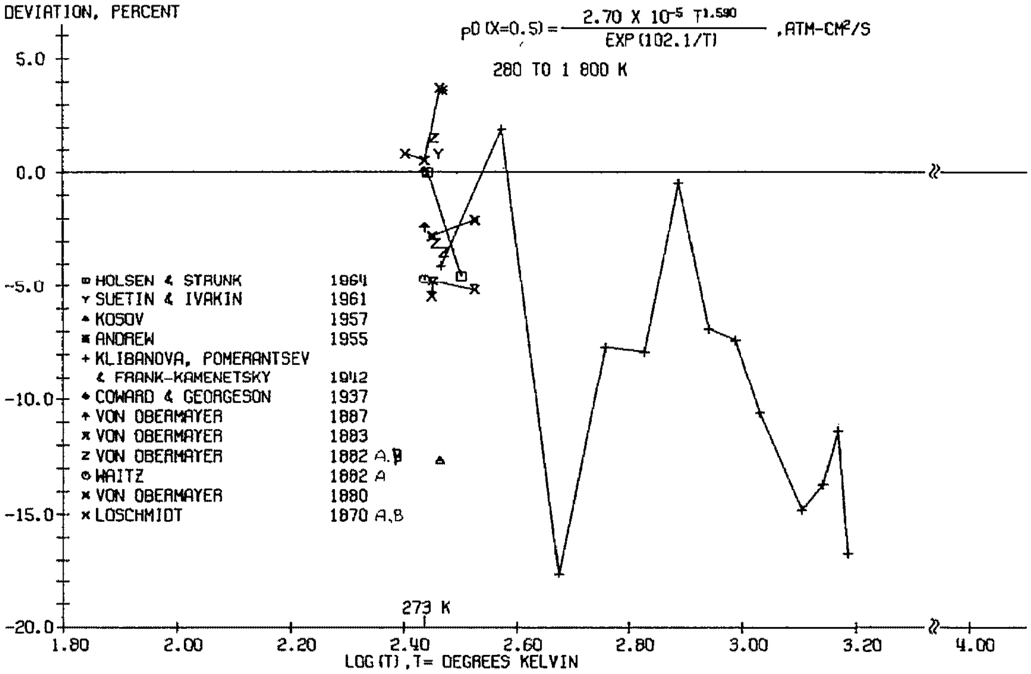

FIGURE 72. *Deviations of diffusion coefficients from reference equation.*

Air—Carbon dioxide

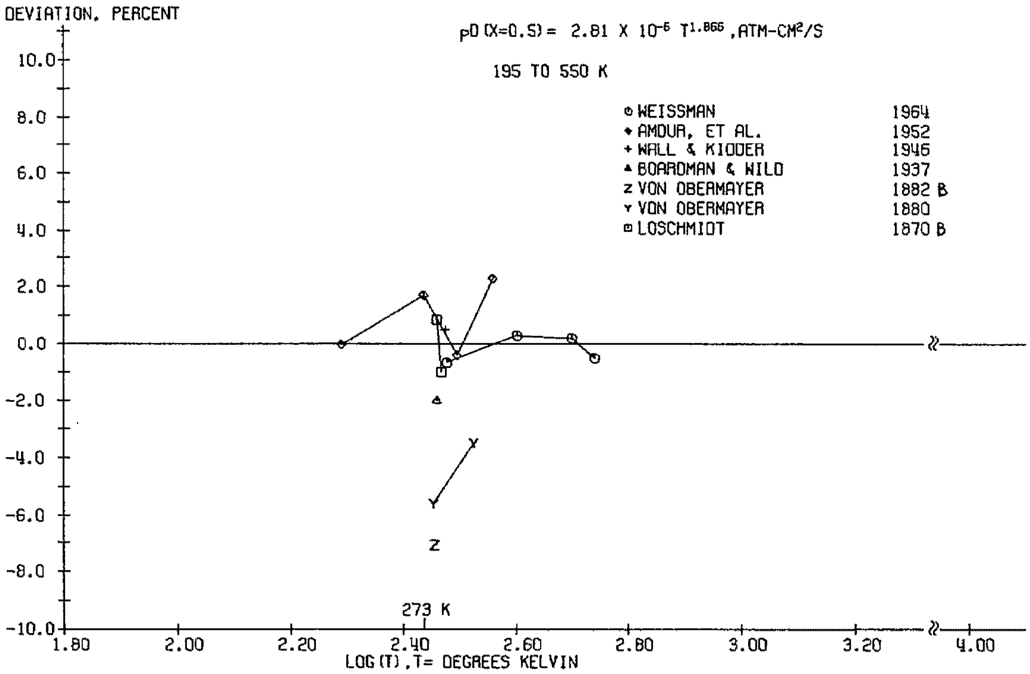

FIGURE 73. *Deviations of diffusion coefficients from reference equation.*

Carbon dioxide—N_2O

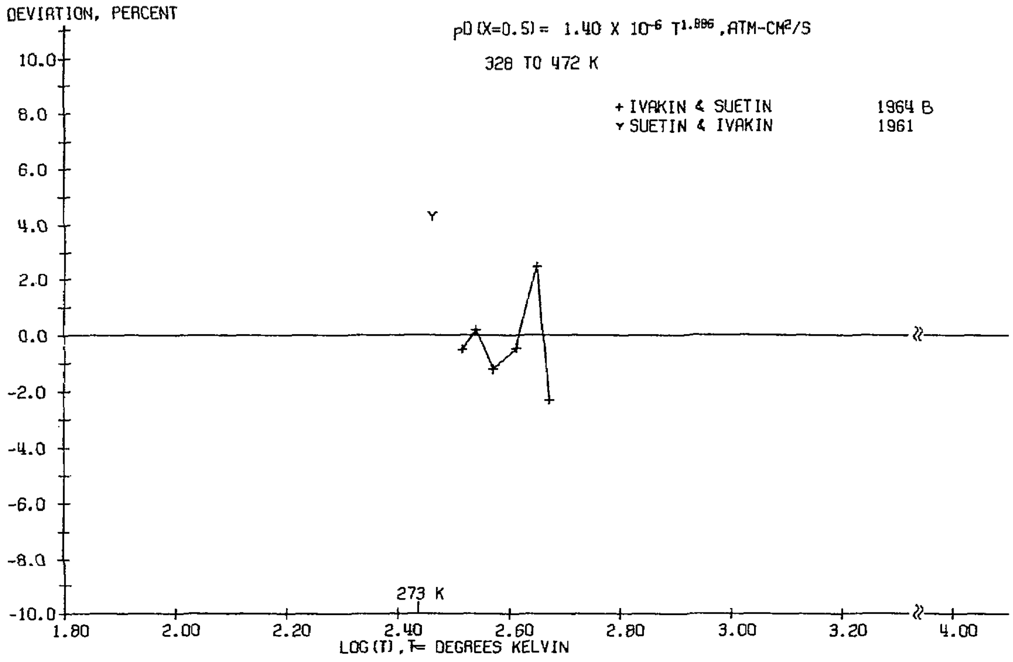

FIGURE 74. *Deviations of diffusion coefficients from reference equation.*

Carbon dioxide–SF_6

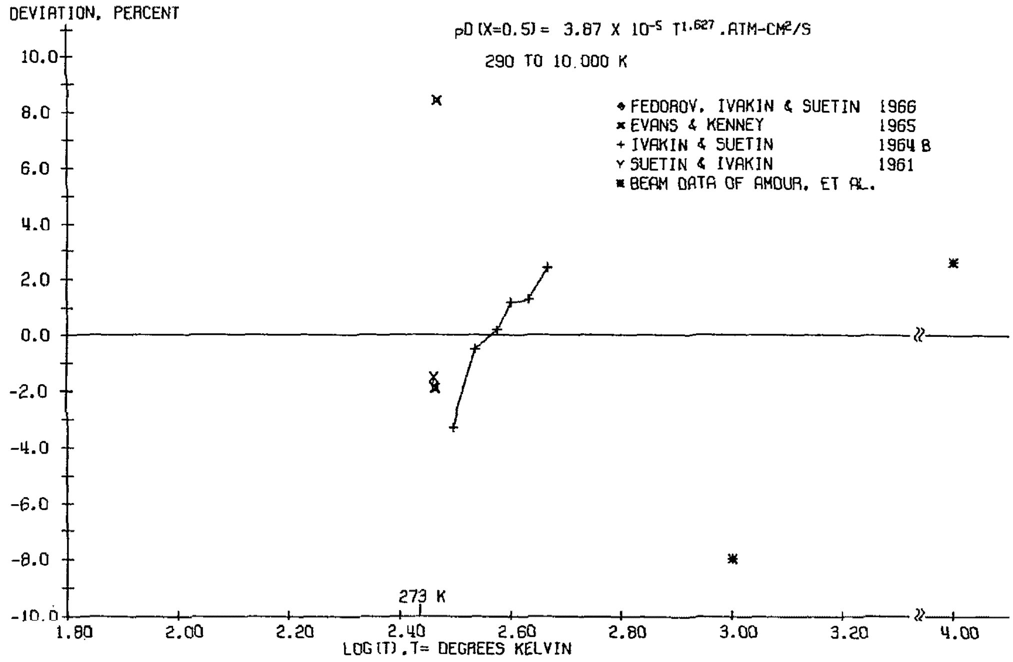

FIGURE 75. *Deviations of diffusion coefficients from reference equation.*

Helium–SF_6

TABLE 22. *Diffusion coefficients and weights for curve-fitting, Group III* [a]

System	T, K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note
Ar-CH_4	307.15	−0.6655	b
	8660	1.9232	c
Ar-CO	—	—	d
Ar-O_2	316	−0.670	*
	3160	1.066	*
Ar-air	282	−0.7520	e
	355	−.5784	e
	447	−.4029	e
	1000	.2095	e
	10 000	1.9576	e
Ar-CO_2	276.2	−0.8775	f
	317.2	−.7820	f
	328.0	−.7328	g
	348.0	−.6819	g
	373.0	−.6289	g
	410.0	−.5528	g
	455.0	−0.4737	g
	473.0	−.4401	g
	1100	.2122	h
	1800	.5752	h
Ar-SF_6	328.0	−1.0000	g
	348.0	−0.9508	g
	373.0	−.8962	g
	410.0	−.8210	g
	447.0	−.7375	g
	472.0	−.6981	g
	1000	−.0985	i
	4640	1.0086	i
H_2-Xe	242.2	−0.3872	j
	274.2	−.2941	j
	303.9	−.2132	j
	341.2	−.1244	j
	293.2	−.2277	k
	400.0	.0065	k
	500.0	.1847	k
	550.0	.2529	k
	2320	1.2989	i
	8290	2.2856	i
H_2-CH_4	316	−0.092	*
	10 000	2.556	*
H_2-O_2	316	−0.050	*
	3160	1.682	*
H_2-SF_6	298.15	−0.3788	l
	286.2	−.4023	m
	306.9	−.3391	m
	370.8	−.1891	m
	418.0	−.0768	m
	313.0	−.3006	g
	344.4	−.2441	g
	376.0	−.1791	g
	401.0	−.1249	g
	429.0	−.0680	g
	473.0	.0128	g
	1320	.7185	i
	7460	1.9782	i
CH_4-He	316	−0.130	*
	3160	1.620	*
CH_4-N_2	316	−0.625	*
	10 000	2.000	*
CH_4-O_2	293.6_5	−0.6676	n
	395	−.4168	n
	402	−.3936	n
	408	−.3990	n
	517	−.2122	n
	521	−.2048	n
	534	−.1993	n
	668	−.0101	n
	669	−.0031	n
	707	.0378	n
	708	.0418	n
	768	.0831	n
	771	.0973	n
	840	.1523	n
	842	.1467	n
	845	.1399	n
	3550	1.250	*
	10 000	2.000	*
CH_4-air	282	−0.7077	e
	355	−.5317	e
	447	−.3551	e
	1000	0.2577	e
	10 000	2.0009	e
CH_4-SF_6	297.58	−0.9566	o
	357.92	−.8119	o
	418.19	−.6872	o
	477.87	−.5784	o
	2045	0.5159	i
	5000	1.0212	i
N_2-Ne	293.15	−0.4989	p
	6090	1.7973	i
N_2-Kr	316	−0.770	*
	5620	1.438	*
N_2-Xe	316	−0.855	*
	7940	1.650	*
N_2-O_2	316	−0.638	*
	3160	1.086	*
N_2-SF_6	328.0	−0.9393	g
	348.0	−.8861	g
	373.0	−.8327	g
	410.0	−.7471	g
	455.0	−.6615	g
	473.0	−.6364	g
	1000	−.0640	i
	3960	.9294	i
CO-Kr	—	—	d
CO-O_2	—	—	d
CO-air	282	−.7077	e
	355	−.5376	e
	447	−.3686	e
	1000	.2279	e
	10,000	1.9727	e
CO-CO_2	281.6_5	−0.8187	q
	293.3_5	−.7854	q
	293.1_5	−.8027	q
	315.4	−.7328	g
	348.0	−.6536	g
	373.0	−.5969	g
	410.0	−.5302	g
	455.0	−.4449	g
	473.0	−.4191	g
CO-SF_6	296.8	−1.0521	g
	315.4	−0.9788	g
	348.0	−.9031	g
	373.4	−.8416	g
	410.0	−.7595	g
	455.0	−.6737	g
	473.0	−.6536	g
	1000	−.0640	i
	3960	0.9294	i
O_2-CO_2	286.9_5	−.8069	q
	287.1_5	−.8097	q
	287.1_5	−.8041	q
	296.5_5	−.7932	r
	287.8	−.8125	s
	296	−.8069	n
	409	−.5287	n
	419	−.5229	n
	430	−.4935	n
	596	−.2457	n
	612	−.2111	n
	635	−.1858	n
	649	−.1838	n
	768	−.0434	n
	770	−.0297	n
	864	0.0302	n
	867	.0546	n
	874	.0492	n
	1080	.2084	n
	1081	.2098	n
	1083	.2049	n
O_2-SF_6	297	−1.0044	t
	317	−0.9626	t
	340	−.8894	t
	379	−.7959	t
	408	−.7144	t
	2930	.658	i
	6310	1.212	i
CO_2-air	282	−0.8300	e
	355	−.6387	e
	501	−.3636	e
	708	−.0992	e
	1000	.1575	e
	1590	.4921	e

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

System	T, K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note	System	T, K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note
CO_2-N_2O	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 $\mathscr{D}_{12}$ are weighted one; except in H_2-SF_6 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 N_2-(gas) is suitable because of isosteric molecules.

[e] Calculated from reference equations for N_2-(gas) and O_2-(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] Strehlow (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).

Tab le. 23. *Molecular-beam potentials*, $\varphi(r)=K/r^s$, *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-Ar	171	6.06	Kamnev and Leonas (1965 a).
Ar-O_2	1360	8.34	2.01 −2.50	Direct measurement			Jordan et al. (1970).
	5000	9.9	2.15 −2.63	Direct measurement			Belyaev and Leonas (1967 a).
Ar-SF_6	24.5×10^5	12.8	3.24 −4.04	He-SF_6	1.86×10^5	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).
	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-CH_4	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).
	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).
H_2-SF_6	4.78×10^5	11.61	3.04 −3.53	He-SF_6	1.86×10^5	11.48	Amdur (1967).
				He-H_2	12.11	6.07	Amdur and Smith (1968).
				He-He	4.71	5.94	Amdur and Harkness (1954).
CH_4-He	602	9.43	1.92 −2.37	Direct measurement			Amdur et al. (1961 b).
CH_4-N_2	832	7.30	2.41 −2.80	Ar-N_2	755	7.78	Amdur et al. (1957).
				Ar-CH_4	936	7.85	Mason and Amdur (1964).
				Ar-Ar	849	8.33	Amdur and Mason (1954).
CH_4-O_2	1500	7.86	2.14 −2.47	Ar-CH_4	936	7.85	Mason and Amdur (1964).
				Ar-O_2	1360	8.34	Jordan et al. (1970).
				Ar-Ar	849	8.33	Amdur and Mason (1954).
CH_4-SF_6	28.0×10^5	12.08	3.54 −3.75	He-SF_6	1.86×10^5	11.48	Amdur (1967).
				Ar-CH_4	936	7.85	Mason and Amdur (1964).
				He-Ar	62.1	7.21	Amdur et al. (1954).
Ne-N_2	605	9.08_5	2.03_5−2.56	He-N_2	74.3	7.06	Amdur et al. (1957).
				He-He	4.71	5.94	Amdur and Harkness (1954).
				Ne-Ne	312	9.99	Amdur and Mason (1955 a).
	207	7.52_5	2.02 −2.61_5	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	432	6.80	2.36_5−3.06_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	
	872	7.55	$2.37-3.03_5$	N_2-N_2	550	7.4	Belvaev and Leonas (1967 a).
				Kr-Kr	1382	7.7	Kamnev and Leonas (1966 a).
N_2-Xe	2874	8.07_5	$2.66-3.29_5$	He-N_2	74.3	7.06	Amdur et al. (1957).
				He-He	4.71	5.94	Amdur and Harkness (1954).
				Xe-Xe	7050	7.97	Amdur and Mason (1956 a).
	505	6.87_5	$2.41-3.07$	N_2-N_2	550	7.4	Belyaev and Leonas (1967 a).
				Xe-Xe	463	6.35	Kamnev and Leonas (1966 a).
N_2-O_2	1630	8.15	$2.16-2.52$	Ar-O_2	1360	8.34	Jordan et al (1970).
				He-N_2	74.3	7.06	Amdur et al. (1957).
				He-Ar	62.1	7.25	Amdur et al. (1954).
	330	6.8	$2.34-3.05$	Direct measurement			Belyaev and Leonas (1967 a).
N_2-SF_6	29.3×10^5	12.60	$3.39-4.06$	He-SF_6	1.86×10^5	11.48	Amdur (1967).
				He-N_2	74.3	7.06	Amdur et al. (1957).
				He-He	4.71	5.94	Amdur and Harkness (1954).
CO-Kr	238	5.53_5	$2.21-2.90_5$	Ar-CO	551	6.99	Jordan et al. (1970).
				Ar-Ar	849	8.33	Amdur and Mason (1954).
				Kr-Kr	159	5.42	Amdur and Mason (1955 b).
	1648	7.96_5	$2.35_5-2.97$	CO-CO	1965	8.23	Belyaev et al. (1967).
				Kr-Kr	1382	7.7	Kamnev and Leonas (1966).
CO-O_2	883	7.00	$1.92-2.49$	CO-Ar	551	6.99	Jordan et al. (1970).
				O_2-Ar	1360	8.34	Jordan et al. (1970).
				Ar-Ar	849	8.33	Amdur and Mason (1954).
	687	7.26_5	$2.32_5-3.03_5$	CO-CO	1965	8.23	Belyaev et al. (1967).
				O_2-O_2	240	6.3	Belyaev and Leonas (1967 a).
CO-SF_6	16.5×10^5	11.22	$3.32-3.77$	He-SF_6	1.86×10^5	11.48	Amdur (1967).
				Ar-CO	551	6.99	Jordan et al. (1970).
				He-Ar	62.1	7.25	Amdur et al. (1954).
O_2-SF_6	40.7×10^5	12.53	$3.24-3.59$	He-SF_6	1.86×10^5	11.48	Amdur (1967).
				Ar-O_2	1360	8.30	Jordan et al. (1970).
				He-Ar	62.1	7.25	Amdur et al. (1954).
He-SF_6	1.86×10^5	11.48	$2.87-3.36$	Direct measurement			Amdur (1967).

[a] Potentials were not determined for air-(Ar, CH_4, CO, SF_6) and CO_2-(Ar, CO, O_2, air, N_2O, SF_6) 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 $\mathscr{D}_{12}$ by molecular-beam measurements with the results for Ar-N_2 (Group II) were the bases for placing Ar-CO into Group III.

Ar-O_2. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$ 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-CO_2. The more reliable measurements for Ar-CO_2 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-SF_6. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$, obtained by the closed-tube method (Ivakin and Suetin, 1964 b).

H_2-Xe. The two-bulb measurements by Paul and Srivastava (1961 c) and the $\mathscr{D}_{12}$ from mixture viscosity data calculated by Weissman and Mason (1962 b) are considered equally reliable.

H_2-CH_4. 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 $\mathscr{D}_{12}$ at temperatures up to 523 K were obtained from mixture viscosity data by Weissman and Mason (1962 b).

H_2-O_2. 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 H_2-N_2 (Group I), which are expected to be similar. Spontaneous ignition occurred at about 920 K in the point-source measurements.

H_2-SF_6. 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).

CH_4-He. The reliable direct measurements of $\mathscr{D}_{12}$ are primarily from recent open-tube studies by Frost (1967) and by Rhodes and Amick (1967).

CH_4-N_2. The two-bulb measurements by Mueller and Cahill (1964) were considered sufficient to place this gas pair into Group III.

CH_4-O_2. 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 CH_4-N_2.

CH_4-air. There are no direct measurements available, and the results were calculated by Blanc's law.

CH_4-SF_6. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$, obtained by the closed-tube method (Manner, 1967).

N_2-Ne. This gas pair has reliable values of $\mathscr{D}_{12}$ only from mixture viscosity measurements by DiPippo et al. (1967).

N_2-Kr. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$, obtained by the two-bulb method (Durbin and Kobayashi, 1962).

N_2-Xe. This gas pair has only one set of direct measurements of $\mathscr{D}_{12}$, obtained by the two-bulb method (Paul and Srivastava, 1961 b).

N_2-O_2. 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.

N_2-SF_6. 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 $\mathscr{D}_{12}$, obtained by the two-bulb method (Singh et al., 1967). Since CO and N_2 are isosteric molecules, the reference equation for N_2-Kr was used for CO-Kr, and the data agree, see figure 65.

CO-O_2. Since CO and N_2 are isosteric molecules, the reference equation for N_2-O_2 was used for CO-O_2. 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-CO_2. Since CO and N_2 are isosteric molecules, the reference equation for N_2-CO_2 (Group II) can be used for CO-CO_2; a slightly more precise correlation, however, is given for CO-CO_2 based only on its direct measurements. The direct measurements for CO-CO_2 are in the temperature range of 282 to 473 K. The use of the N_2-CO_2 reference equation will extend the higher temperature limit to 1800 K, which is a significant advantage.

CO-SF_6. This gas pair has direct measurements of $\mathscr{D}_{12}$ which were obtained by the closed-tube method (Ivakin and Suetin, 1964 a, b).

O_2-CO_2. 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 $\mathscr{D}_{12}$ (Loschmidt, 1870 b; Wretschko, 1870).

O_2-SF_6. This gas pair has reliable closed-tube measurements by Ivakin et al. (1968), which, however, probably have somewhat too great a temperature dependence for $\mathscr{D}_{12}$ over 297 to 408 K.

CO_2-air. Even though there are many direct measurements available, the reference equation for CO_2-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 Klibanova 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 N_2-CO_2, and by Walker and Westenberg (1960) for O_2-CO_2.

CO_2-N_2O. 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 $\mathscr{D}_{12}$ from mixture viscosity were used to extend the temperature range to 550 K. The placement of CO_2-N_2O into Group III, and not Group II, was decided upon because of the limited temperature range of the direct measurements and the uncertainties in $\mathscr{D}_{12}$ obtained from mixture viscosity for cylindrical molecules.

CO_2-SF_6. This gas pair has reliable closed-tube measurements by Ivakin and Suetin (1964 b).

SF_6-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.

SF_6-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 $\mathscr{D}_{12}$ and their weights used in the least-squares calculations are presented in table 24. Except for mixtures with dissociated gases the values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$. 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 $\mathscr{D}_{12}$ 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.

H_2O-N_2. 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 H_2O diffused downwards through N_2. In the other studies the H_2O was located below the N_2, or the lighter component below the heavier, which would have possible adverse effects due to convection. Because values of $\mathscr{D}_{12}$ are available only from evaporation-tube studies, it may be noted that the temperature range is limited, 282 to 373 K.

H_2O-O_2. The correlation consists of two power functions ($\mathscr{D}_{12}=AT^s$), 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 H_2O-O_2 evaporation-tube studies are considered too uncertain. But, at low temperatures, data by O'Connell et al. (1969) for H_2O-N_2 were systematically increased by 1 percent to obtain values of $\mathscr{D}_{12}$ for H_2O-O_2. This slight adjustment was based on calculations for transport properties of polar-gas mixtures [3]. But all results for H_2O-O_2 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

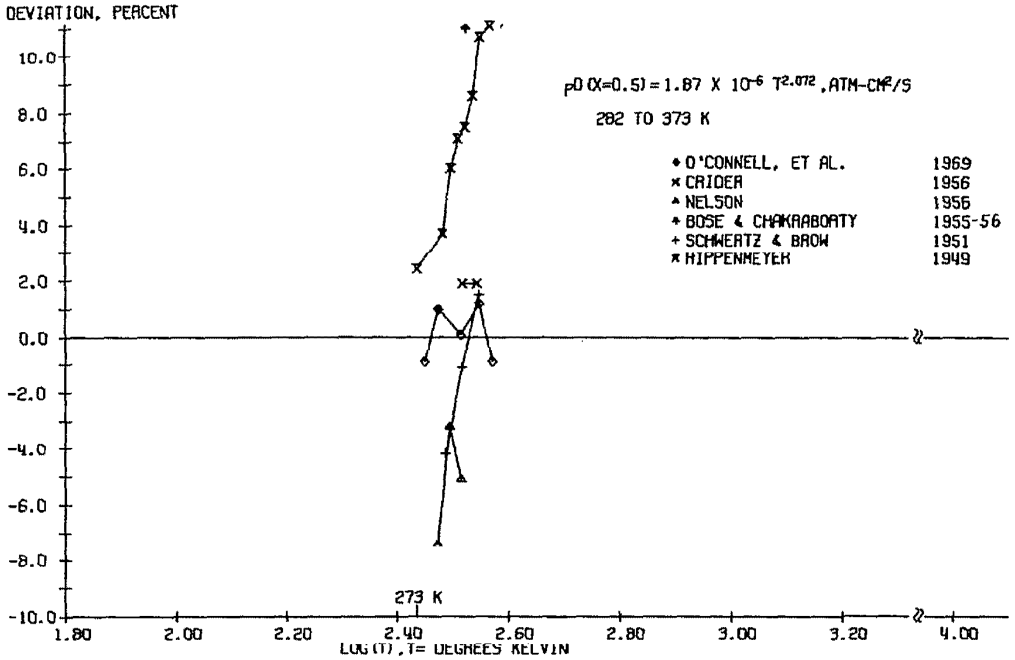

FIGURE 76. *Deviations of diffusion coefficients from reference equation.*

Water–Nitrogen

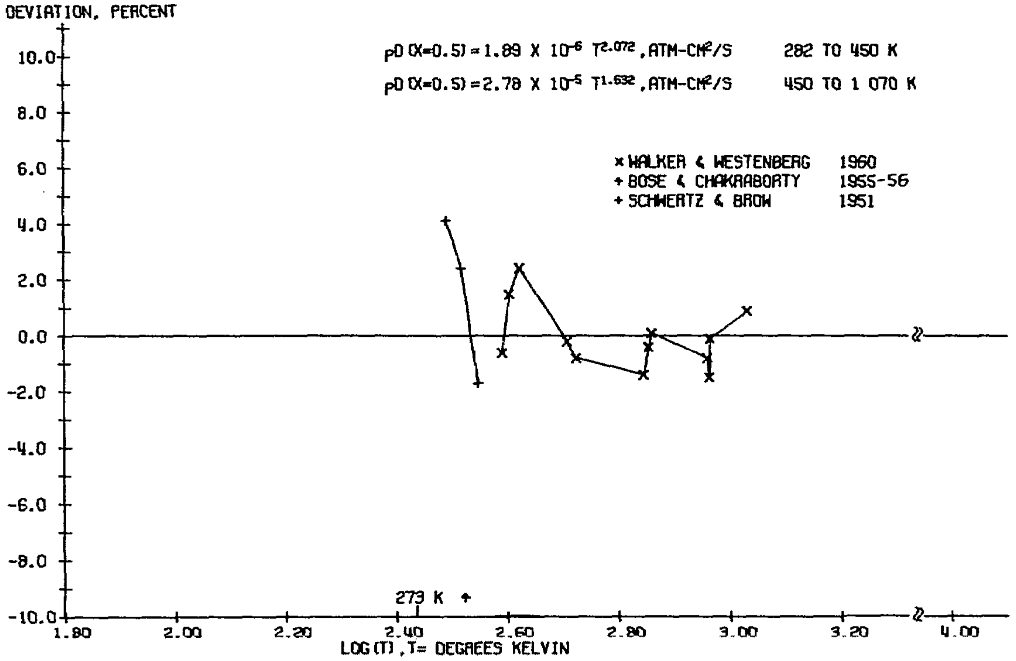

FIGURE 77. *Deviations of diffusion coefficients from reference equation.*

Water–Oxygen

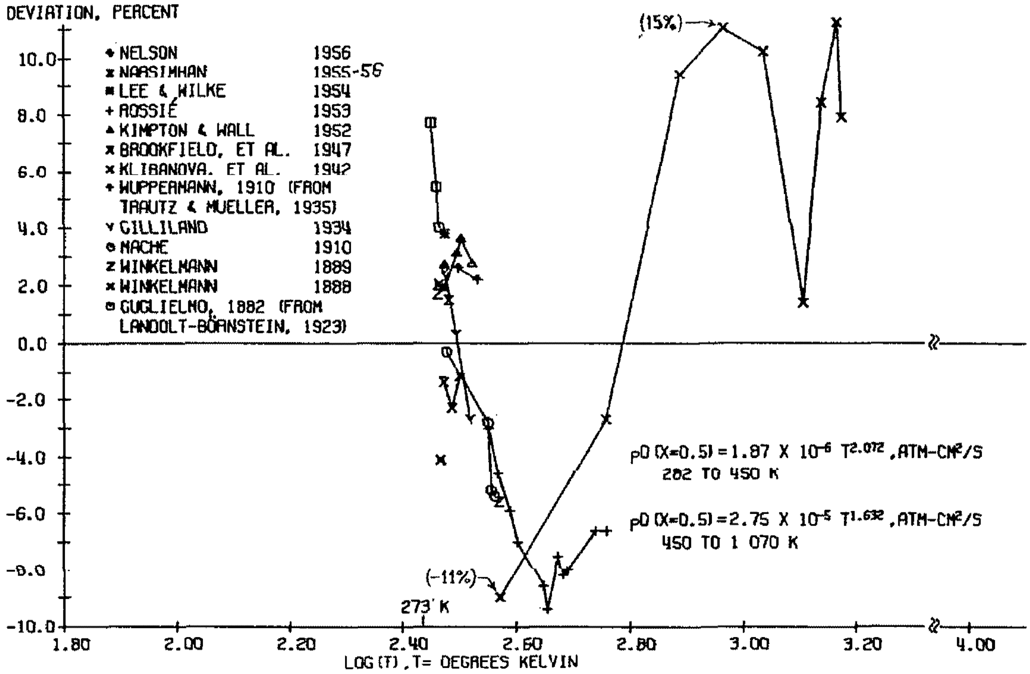

FIGURE 78. *Deviations of diffusion coefficients from reference equation.*

Water—Air

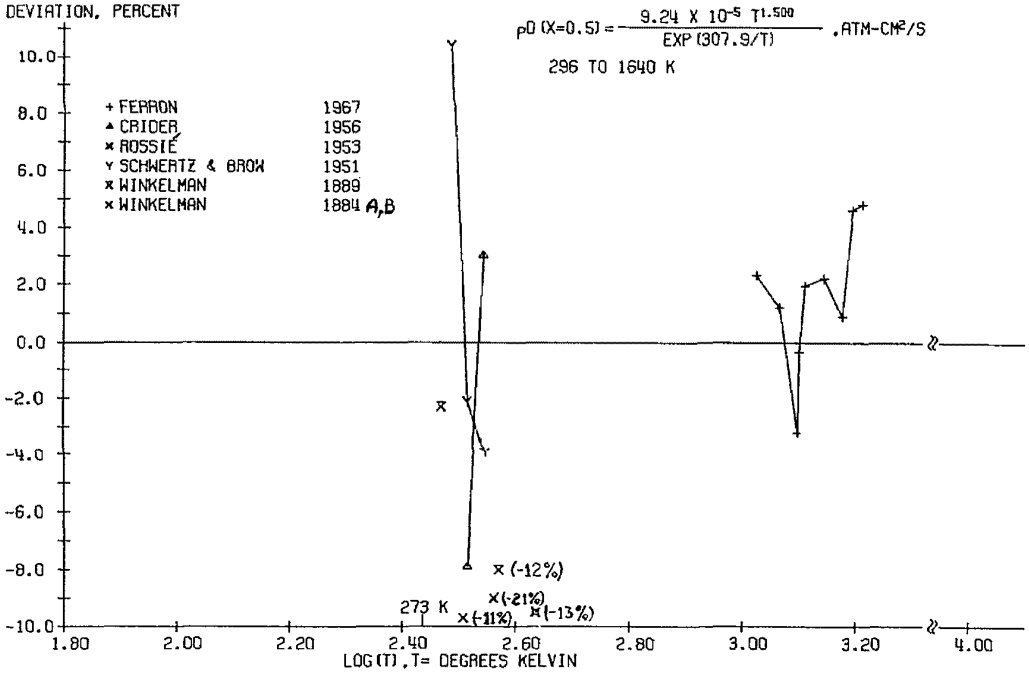

FIGURE 79. *Deviations of diffusion coefficients from reference equation.*

Water—Carbon dioxide

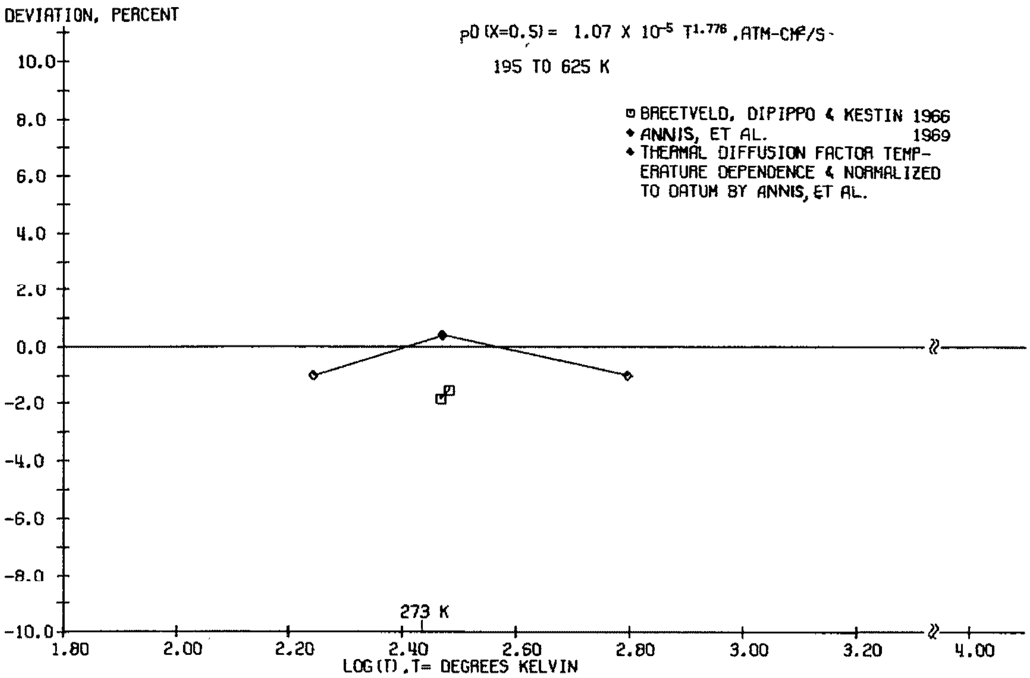

FIGURE 80. *Deviations of diffusion coefficients from reference equation.*

Neon—Carbon dioxide

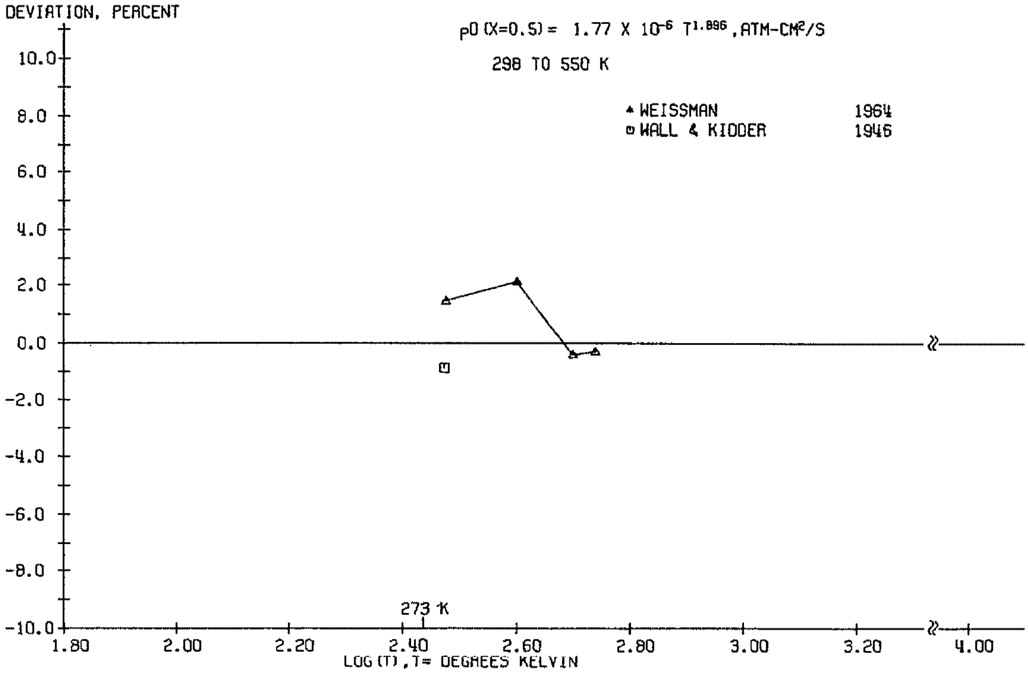

FIGURE 81. *Deviations of diffusion coefficients from reference equation.*

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.

H_2O-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 H_2O-N_2 and H_2O-O_2 have too much scatter, and a verification of a correlation by Blanc's law from direct measurements for H_2O-air would not be significant. The reference equation for H_2O-air is based on the synthesis of results by O'Connell et al. (1969) for H_2O-N_2 and by Walker and Westenberg (1960) for H_2O-O_2. Since air is approximately 80 percent N_2, the reference equation for H_2O-N_2 extrapolated to 450 K, was assumed directly applicable at low temperatures. For temperatures between 450 and 1070 K, the reference equation for H_2O-O_2 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 H_2O-air, which are listed in table 16.

H_2O-CO_2. 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.

CO_2-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 $\mathscr{D}_{12}$ between 195 to 625 K.

CO_2-C_3H_8. The closed-tube measurements by Wall and Kidder (1946) and the values of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}=AT^s$. 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note	System	T, K	$\log_{10}[\mathscr{D}_{12}(x=1/2)]$	Note
H_2O-N_2	281.9	−0.6554	b	H_2O-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
	353.2	−.4436	b		1700	.7482	h
	373.4	−.4029	b	CO_2-Ne	175	−.9914	i
H_2O-O_2	450.0	−.2261	c		625	−.0097	i
	511	−.1367	d	CO_2-C_3H_8	298.1_5	−1.0665	j
	529	−.1146	d		298.1_5	−1.0620	j
	698	+.0792	d		300.0	−1.0492	k
	715	.1007	d		400.0	−0.8097	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_2O-air	—	—	e		10 000	2.966	*
H_2O-CO_2	307.4_5	−.6947	f	N-N_2 O-N_2 O-O_2	280	−0.5376	n
	328.5_5	−.6757	f		2850	1.250	*, n
	352.3_5	−.6108	f				
	328.6_5	−.7033	g	O-He	316	0.043	*
	349.1_5	−.5901	g		10 000	2.6665	o
	1000	.3617	h	O-Ar	316	−0.522	*
	1100	.4099	h		3760	1.457	*

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

[a] All listed values of $\mathscr{D}_{12}$ are weighted one; except in H_2O-CO_2 for which the data between 1000 and 1700 K inclusive, are weighted two. For these systems, in almost all instances, the composition dependence of $\mathscr{D}_{12}$ is insignificant; maximum correction is 0.45 percent for H_2O-O_2 datum at 1070 K.

[b] O'Connell et al. (1969).

[c] Common point for low- and high-temperature correlations, see text (Section 5.4, part d).

[d] Walker and Westenberg (1960, 1966).

[e] See section 5.4, part d.

[f] Schwertz and Brow (1951).

[g] Crider (1956).

[h] Ferron (1967); weight of datum doubled for least-squares calculations.

[i] Calculated 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.

[j] Wall and Kidder (1946).

[k] Weissman (1964).

[l] Khouw et al. (1969).

[m] Browning and Fox (1964).

[n] Morgan and Schiff (1964), average of results for N-N_2, O-N_2, and O-O_2.

[o] Calculated from potential of Leonas et al., see table 25.

TABLE 25. *Molecular-beam potentials,* $\varphi(r) = K/r^s$, *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).
				He-Ar	22.6	5.15	Kamnev and Leonas (1965 a).
				He-He	4.33	5.86	Belyaev and Leonas (1967 b).
$H-H_2$	6.02	3.42	1.33–1.88	He-H	2.34	3.29	Amdur and Mason (1956 b).
				$He-H_2$	12.11	6.07	Amdur and Smith (1968).
				He-He	4.71	5.94	Amdur and Harkness (1954).
	0.91	4.15	1.00–1.24	Direct measurement			Belyaev and Leonas (1967 b).
$N-N_2$	76.6	6.31	1.76–2.54	Direct measurement			Belyaev and Leonas (1966 b).
O-He	38.0	7.99	1.20–1.60	Ar-O	239	8.09	Belyaev et al. (1967).
				He-He	4.33	5.86	Belyaev and Leonas (1967 b).
				Ar-Ar	171	6.06	Kamnev and Leonas (1965 a).
O-Ar	239	8.09	1.78–2.40	Direct measurement			Belyaev et al. (1967).
$O-N_2$	22.5	5.0	2.00–2.48	Direct measurement			Belyaev and Leonas (1966 b).
$O-O_2$	13.25	4.4	2.05–2.46	Direct measurement			Belyaev and Leonas (1966 b).

[a] Potentials were not determined for H_2O-(N_2., O_2, air, CO_2) and CO_2-(N_2O, C_3H_8) because molecular-beam measurements were unavailable.
[b] Complete reference information is given in Bibliography II.

H-H_2. For this gas pair the most reliable values of $\mathscr{D}_{12}$ are considered to be from mixture viscosity (Browning and Fox, 1964). The other determinations of $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ 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 $\mathscr{D}_{12}$ which are too high in comparison with both the low-temperature data, and results of other beam measurements.

N-N_2, O-N_2, O-O_2. For these gas pairs, with similar diffusion characteristics, the results by Morgan and Schiff (1964) are considered to be the most reliable. For O-O_2, 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 $\mathscr{D}_{12}$ from molecular-beam measurements for N-N_2, O-N_2, O-O_2 (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 $\mathscr{D}_{12}$ at elevated temperatures were based on a single laboratory source for the potentials (Leonas et al.).

TABLE 16. *Experimental determinations of* $\mathscr{D}_{12}$ *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
^3He-^3He	Luszczynski et al. (1962)	1.7 to 4.2
	Luszczynski et al. (1967)	1.13 to 4.22
^3He-^4He	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
^4He-^4He	Amdur and Mason (1958)	1000 to 15 000
He-Ne	Srivastava and Barua (1959)	273 to 318
	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* $\mathscr{D}_{12}$ *according to gas pair, temperature noted*—Continued

a. Noble Gases—Continued

System	Reference	T, K
	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
He-Ar	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)	273 to 318
	Weissman and Mason (1962 b)	291 to 550
	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 1000
He-Rn	Hirst and Harrison (1939)	283–286
Ne-Ne	Groth and Sussner (1944)	293
	Winn (1950)	78 to 353
	Amdur and Mason (1958)	1000 to 15 000
Ne-Ar	Schäfer and Schuhmann (1957)	90 to 473

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

a. Noble Gases—Continued

System	Reference	T, K
	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
Ne-Kr	Srivastava and Srivastava (1959)	273 to 318
	Paul (1962)	302
	Weissman and Mason (1962 b)	291
	Watts (1964)	303
	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
Ne-Xe	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
	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
Ne-Rn	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* $\mathscr{D}_{12}$ *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
	van Heijningen et al. (1968)	169 to 400
Xe-Xe	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
He-TH	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
	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
He-NH_3	Giddings and Seager (1962)	297
	Srivastava (1962)	274 to 333
	Ivakin and Suetin (1964 a)	297
He-H_2O	Schwertz 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
	Gozzini et al. (1967)	273 and 443
	Violino (1968)	427 to 443
He-C_2H_2	Suetin and Ivakin (1961)	290
	Suetin (1964)	290
He-C_2H_4	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
	Arai et al. (1967)	313
He-C_2H_6	Frost (1967)	303 to 751
	Kaufmann (1967)	373 to 523
He-O_2	Paul and Srivastava (1961 a)	244 to 334
	Suetin and Ivakin (1961)	287

TABLE 16. *Experimental determinations of* $\mathscr{D}_{12}$ *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
	Wasik and McCulloh (1969)	298 to 365
He-air	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
	Evans et al. (1969)	295
He-CH_3OH	Seager et al. (1963)	423 to 523
He-K	Bernheim and Korte (1965)	358
	Khomchenkov et al. (1968)	723
	Violino (1968)	358
	Ivanovskiĭ et al. (1969)	570 to 930
He-C_3H_6	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_3H_8	Frost (1967)	303 to 767
	Kaufmann (1967)	373 to 503
	Rhodes and Amick (1967)	303
He-C_2H_5OH	Lee and Wilke (1954)	298
	Seager et al. (1963)	423 to 523
	Hargrove and Sawyer (1967)	298
He-difluoromethane	Fuller et al. (1969)	431
He-1-butene	Frost (1967)	303 to 522
He-2-butene	Weissman (1964)	293 to 523
He-C_4H_{10}	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 $\mathscr{D}_{12}$ according to gas pair, temperature noted*—Continued

b. Noble Gases and Another Component—Continued

System	Reference	T, K
He-*n*-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
	Khomchenkov 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 $\mathscr{D}_{12}$ 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
	Violino (1968)	(?)
Ar-C_2H_2	Suetin and Ivakin (1961)	287
	Suetin (1964)	287
	Ivakin et al. (1968)	298 to 407
Ar-C_2H_4	Weissman (1964)	298
Ar-N_2	Waldmann (1944, 1947)	293
	Schäfer and Moesta (1954)	233 to 363
	Westenberg and Walker (1957)	293
	Paul and Srivastava (1961 b)	244 to 335
	DiPippo et al. (1967)	293 and 303
Ar-CO	Ivakin and Suetin (1964 a)	296
Ar-C_2H_6	Jacobs et al. (1970)	298
Ar-O_2	Waldmann (1944, 1947)	293
	Paul and Srivastava (1961 a)	243 to 334
	Scott and Dullien (1962)	293
Ar-air	Evans et al. (1969)	295
Ar-K	Khomchenkov et al. (1968)	723
	Ivanovskiĭ et al. (1969)	630 to 950
Ar-C_3H_8	Jacobs et al. (1970)	298
	Lannus and Grossman (1970 a, b)	242 to 473
Ar-CO_2	Waldmann (1944, 1947)	293
	Suetin and Ivakin (1961)	289
	Ivakin and Suetin (1964 b)	289 to 473
	Holsen and Strunk (1964)	276 and 317
	Suetin (1964)	289
	Pakurar and Ferron (1964)	295, and 1181 to 1676
	Kestin et al. (1966)	293 and 303
	Kosov and Novosad (1966 a)	294
	Pakurar and Ferron (1966)	1132 to 1798
	Ferron (1967)	1100 to 1800
	Oost et al. (1967)	295 and 343
	Gurvich and Matizen (1968)	308
Ar-Cr	Grieveson and Turkdogan (1964)	1600
Ar-Fe	Grieveson and Turkdogan (1964)	1600
Ar-acetone	Hargrove and Sawyer (1967)	298 to 473
Ar-*n*-butane	Hargrove and Sawyer (1967)	298 to 473
	Jacobs et al. (1970)	298
Ar-Ni	Grieveson and Turkdogan (1964)	1600
Ar-Co	Grieveson and Turkdogan (1964)	1600
Ar-nitromethane	Byrne et al. (1967)	303
Ar-SO_2	Schäfer (1959)	263
Ar-Zn	Nikolaev and Aleskovskiĭ (1964)	1100 to 2600
Ar-*n*-pentane	Hargrove and Sawyer (1967)	298 to 473
Ar-ether	Hargrove and Sawyer (1967)	298 to 473
Ar-benzene	Hargrove and Sawyer (1967)	298 to 473
Ar-HBr	Mian (1967)	328 to 523
	Mian et al. (1969)	328 to 523
Ar-cyclohexane	Cummings and Ubbelohde (1953, 1955)	289
Ar-methylcyclopentane	Cummings and Ubbelohde (1953, 1955)	289
Ar-Rb	Franzen (1959)	320
	Violino (1968)	320 and 340
Ar-*n*-hexane	Cummings and Ubbelohde (1953, 1955)	289
	Hargrove and Sawyer (1967)	298 to 473
Ar-2,3 dimethylbutane	Cummings and Ubbelohde (1953, 1955)	289
Ar-3-pentanone	Barr and Sawyer (1964)	300
Ar-toluene	Fairbanks and Wilke (1950)	294
Ar-monofluorobenzene	Byrne et al. (1967)	303
Ar-2,4-dimethylpentene	Clarke and Ubbelohde (1957)	303
Ar-*n*-heptane	Clarke and Ubbelohde (1957)	303
Ar-*n*-octane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
Ar-2,2,4-trimethylpentane	Cummings and Ubbelohde (1953, 1955)	303
	Clarke and Ubbelohde (1957)	303
Ar-Cs	Legowski (1964)	299
	Khomchenkov et al. (1968)	723
	Violino (1968)	299 and 317
Ar-SF_6	Suetin and Ivakin (1961)	287
	Suetin (1964)	287
	Ivakin and Suetin (1964 b)	287 to 472
	Evans and Kenney (1965)	293
Ar-1-bromo-3-methylbutane	Byrne et al. (1967)	303
Ar-Br_2	Mackenzie and Melville (1933)	289
Ar-CF_3I	Belousova et al. (1970)	300
Ar-Hg	Spencer et al. (1969)	459 to 607
Ar-C_3F_7I	Belousova et al. (1970)	300
Ar-UF_6	Ljunggren (1965)	293
Ar-As_4	Krol et al. (1967)	853 to 913
Kr-H_2	Miller and Carman (1961)	293
	Mason et al. (1964 b)	296

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

b. Noble Gases and Another Component—Continued

System	Reference	T, K
	Miller and Carman (1964)	293
	Fedorov et al. (1966)	291
	Annis et al. (1968)	77 to 760
	Annis et al. (1969)	295
Kr-D_2	Mason et al. (1964 b)	255 to 362
	Annis et al. (1969)	295
Kr-T_2	Mason et al. (1964 b)	252 to 346
	Annis et al. (1969)	295
Kr-NH_3	Srivastava and Srivastava (1962)	255 to 333
Kr-C_2H_4	Durbin and Kobayashi (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
	Kestin and Yata (1968)	293 and 303
Kr-acetone	Srivastava and Saran (1966 a)	284 to 313
Kr-SO_2	Saran and Singh (1966)	303
	Srivastava and Saran (1966 b)	274 to 318
Kr-C_2H_5Cl	Singh and Srivastava (1968)	275 to 318
Kr-$(C_2H_5)_2O$	Srivastava and Saran (1966 b)	274 to 318
Kr-CH_2Cl_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
	Miller and Carman (1964)	293
Xe-NH_3	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_3I	Belousova et al. (1970)	300
Xe-Hg	Nakayama (1968)	301
Xe-C_3F_7I	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	T, 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
	Sancier and Wise (1969)	293 to 719
N-N_2	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
	Yolles and Wise (1968)	298
	Yolles et al. (1970)	298 to 873

TABLE 16. *Experimental determinations of* $\mathscr{D}_{12}$ *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
	Annis et al. (1969)	295
H_2-HD	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
	Diller and Mason (1966)	14 to 293
H_2-DT	Reichenbacher et al. (1965)	297
H_2-T_2	Amdur and Beatty (1965)	195 to 353
	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_2O	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
	Schwertz 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
	Violino (1968)	473
H_2-C_2H_2	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
	Schneider 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 $\mathscr{D}_{12}$ according to gas pair, temperature noted*—Continued

d. Other Mixtures—Continued

System	Reference	T, K
	Ivakin and Suetin (1964 b)	296 to 471
H_2-C_2H_4	von Obermayer (1883)	287
	Weissman (1964)	195 to 523
H_2-NO	Weissman (1964)	273
H_2-C_2H_6	von Obermayer (1883)	288
	Boyd et al. (1951)	298
	Fejes and Czárán (1961)	298
	Weissman (1964)	293 to 523
H_2-O_2	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
	Zhalgasov and Kosov (1968)	103 to 298
	Saxena and Gupta (1970)	313 to 366
H_2-air	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	Ivanovskiĭ et al. (1969)	680 to 830
H_2-C_3H_6	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 Zhalgasov (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
	Weissman (1964)	273 to 550
H_2-formic acid	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-n-butane	Strehlow (1953)	288 to 430
	Fejes and Czárán (1961)	298
H_2-acetic acid	Winkelmann (1885)	339 to 372

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

d. Other Mixtures—Continued

System	Reference	*T*, K
H_2-*n*-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-*n*-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-*n*-butyl alcohol	Winkelmann (1885)	372
	Huang et al. (1968)	393 to 483
H_2-*i*-butyl alcohol	Winkelmann (1885)	340 and 357
H_2-*sec*-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-*n*-hexane	Cummings and Ubbelohde (1953, 1955)	289
	Huang et al. (1968)	353 to 453
H_2-*n*-butyric acid	Winkelmann (1885)	372
H_2-*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-*n*-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-*n*-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*-butyrate	Winkelmann (1884 c)	323 and 340
H_2-*i*-valeric acid	Winkelmann (1885)	372
H_2-*n*-hexyl alcohol	Winkelmann (1885)	372
H_2-*n*-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*-butyl acetate	Winkelmann (1884 c)	340 and 371
H_2-ethyl butyrate	Winkelmann (1884 c)	340 and 370
H_2-ethyl *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_2Cl_2	Miller and Carman (1961, 1964)	293
H_2-*n*-nonane	Cummings et al. (1955)	340
H_2-*i*-butyl propionate	Winkelmann (1884 c)	371
H_2-propyl butyrate	Winkelmann (1884 c)	371
H_2-propyl *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-*n*-decane	Cummings and Ubbelohde (1953, 1955)	364
H_2-amyl propionate	Winkelmann (1884 c)	371

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

d. Other Mixtures—Continued

System	Reference	T, K
H_2-*i*-butyl butyrate	Winkelmann (1884 c)	371
H_2-*i*-butyl *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*-butyrate	Winkelmann (1884 c)	371
H_2-*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-*n*-dodecane	Cummings and Ubbelohde (1953, 1955)	400
H_2-Hg	Gaede (1915)	Room temperature
	Spier (1939)	314 to 325
H_2-di-*n*-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-*n*-heptane	Clarke and Ubbelohde (1957)	303
D_2-2:4-dimethylpentane	Clarke and Ubbelohde (1957)	303
D_2-*n*-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
CH_4-NH_3	Weissman (1964)	288
CH_4-H_2O	Schwertz and Brow (1951)	308 to 352
	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_3T	Hu and Kobayashi (1970)	298
CH_4-C_2H_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_2H_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_3H_8	Weissman (1964)	293 to 523
	Chang (1966)	255 to 311
CH_4-CH_3Cl	Manner (1967)	298 to 478

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

d. Other Mixtures—Continued

System	Reference	T, K
CH_4-n-C_4H_{10}	Chang (1966)	*255 to 311*
	Kestin and Yata (1968)	293 and 303
	Beatty (1969)	303
CH_4-Rb	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_3T-CF_4	Hu and Kobayashi (1970)	298
CD_3H-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_2H_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
NH_3-CH_3NH_2	Burch and Raw (1967)	273 to 673
NH_3-O_2	Weissman (1964)	293 to 473
NH_3-ethyl ether	Srivastava and Srivastava (1963)	288 to 338
	Pal and Bhattacharyya (1969)	299 to 373
NH_3-SF_6	Ivakin and Suetin (1964 a)	297
H_2O-H_2O	Ferron (1967)	950 to 1400
H_2O-N_2	Hippenmeyer (1949)	273 to 368
	Schwertz 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
H_2O-C_2H_4	Schwertz and Brow (1951)	308 to 353
	Kimpton and Wall (1952)	298
H_2O-C_2H_6	Kimpton and Wall (1952)	298
	Bose and Chakraborty (1955–56)	332 and 336
H_2O-O_2	Schwertz and Brow (1951)	308 to 352
	Walker and Westenberg (1960, 1966)	390 to 1070
H_2O-air	Stefan (1871)	Room temperature
	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
H_2O-CH_3OH	Weissman (1968 a)	373
H_2O-H_2O_2	Weissman (1968 a)	443 to 513
H_2O-CO_2	Guglielmo (1882)	291
	Winkelmann (1884 a, b)	323 and 366
	Winkelmann (1889)	294–298 and 373
	Trautz and Müller (1935)	294 to 372
	Schwertz and Brow (1951)	307 to 352
	Rossié (1953)	433
	Crider (1956)	329 and 349

TABLE 16. *Experimental determinations of* $\mathscr{D}_{12}$ *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_2O-C_3H_8	Kimpton and Wall (1952)	298
H_2O-C_2H_5OH	Weissman (1968 a)	373
H_2O-SO_2	Kimpton and Wall (1952)	298
H_2O-ethyl ether	Winkelmann (1884 a)	284 and 293
H_2O-CCl_2F_2	Lee and Wilke (1954)	298
D_2O-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
Na-C_5H_{12}	von Hartel et al. (1932)	655
C_2H_2-C_2H_2	Mueller and Cahill (1964)	298 to 383
C_2H_2-C_2H_4	Mueller and Cahill (1964)	298 to 383
C_2H_2-C_2H_6	Mueller and Cahill (1964)	298 to 383
C_2H_2-O_2	Suetin and Ivakin (1961)	287
	Suetin (1964)	287
C_2H_2-air	Kosov (1957)	289
	Suetin and Ivakin (1961)	288
	Suetin (1964)	288
C_2H_2-C_3H_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_2H_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_2H_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
	Lonius (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_3OH	Arnikar and Ghule (1969)	355
N_2-HCl	Mian (1967)	324 to 523
	Mian et al. (1969)	324 to 523
N_2-K	Ivanovskiĭ 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 $\mathscr{D}_{12}$ 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
	Lannus and Grossmann (1970 a, b)	283 to 472
N_2-C_2H_5OH	Bose and Chakraborty (1955–56)	327 and 331
	Arnikar et al. (1967 b)	353
	Arnikar and Ghule (1969)	355
N_2-acetone	Mehta (1966)	298
	Arnikar et al. (1967 b)	353
	Arnikar and Ghule (1969)	353
	Nagata and Hasegawa (1970)	343 to 383
N_2-*n*-butane	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*-butane	Boyd et al. (1951)	298
N_2-*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-*n*-pentane	Arnikar and Ghule (1969)	353
N_2-*n*-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-*n*-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-*n*-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* $\mathscr{D}_{12}$ *according to gas pair, temperature noted*—Continued

d. Other Mixtures—Continued

System	Reference	T, K
N_2-*n*-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-*n*-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-*n*-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-*n*-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
	Weissman (1964)	300 to 550
CO-O_2	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_2	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-*n*-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* $\mathscr{D}_{12}$ *according to gas pair, temperature noted*—Continued

d. Other Mixtures—Continued

System	Reference	T, K
O_2-2:3-dimethylbutane	Cummings and Ubbelohde (1953, 1955)	288
O_2-*n*-hexane	Cummings and Ubbelohde (1953, 1955)	289
O_2-tetrahydrothiophen	Hudson et al. (1960)	319
O_2-*n*-octane	Cummings and Ubbelohde (1953, 1955)	303
O_2-2:2:4-trimethypentane	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_3OH	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_2O_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_2H_5OH	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
air-allyl alcohol	Lugg (1968)	298
air-acetic acid	Winkelmann (1885)	339 to 372
	Pochettino (1914)	336 to 372
	Lugg (1968)	298
air-methyl formate	Griboiedov (1893)	289–295
	Pochettino (1914)	284 and 293
	Lugg (1968)	298
air-*n*-propyl alcohol	Winkelmann (1885)	340 and 357
	Vaillant (1911)	287
	Pochettino (1914)	288 to 355
	Lugg (1968)	298
air-*i*-propyl alcohol	Pochettino (1914)	288 and 333
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air-ethylene diamine	Lugg (1968)	298
air-CNCl	Klotz and Miller (1947)	273
air-ethylene glycol	Lugg (1968)	298
air-SO_2	Andrew (1955)	293

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

d. Other Mixtures—Continued

System	Reference	T, K
air-2-methyl-1,3-butadiene	Altshuller and Cohen (1960)	288
air-Cl_2	Andrew (1955)	293
	Kosov (1957)	289
air-methylethylketone	Lugg (1968)	298
air-pentane	Lugg (1968)	298
air-dimethylformamide	Lugg (1968)	298
air-*n*-butylamine	Pochettino (1914)	334–335
	Lugg (1968)	298
air-*i*-butylamine	Pochettino (1914)	292 and 335
	Lugg (1968)	298
air-diethylamine	Pochettino (1914)	283 and 324
	Lugg (1968)	298
air-methyl acetate	Winkelmann (1884 c)	294 and 319
	Griboiedov (1893)	300 to 317
	Pochettino (1914)	283 to 324
	Lugg (1968)	298
air-ethyl formate	Winkelmann (1884 c)	294 and 319
	Pochettino (1914)	284 to 323
	Lugg (1968)	298
air-propionic acid	Winkelmann (1885)	366 and 372
	Pochettino (1914)	324 to 372
	Lugg (1968)	298
air-*n*-butyl alcohol	Winkelmann (1885)	372
	Pochettino (1914)	334 and 373
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air-*i*-butyl alcohol	Winkelmann (1885)	340 and 357
	Pochettino (1914)	333 and 372
	Lugg (1968)	298
air-*sec*-butyl alcohol	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air-*tert*-butyl alcohol	Pochettino (1914)	294 and 340
	Lugg (1968)	298
air-ethyl ether	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
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
	Heinzelmann 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(?)
air-ethylene chlorohydrin	Lugg (1968)	298
air-cyclohexane	Goryunova and Kuvshinskiĭ (1948)	318
air-hexene	Altshuller and Cohen (1960)	293 and 303
air-dichloromethane	Lugg (1968)	298
air-methylpropylketone	Lugg (1968)	298
air-*n*-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
	Grob and El-Wakil (1969)	297
air-*n*-butyric acid	Winkelmann (1885)	372

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

d. Other Mixtures—Continued

System	Reference	*T*, K
	Pochettino (1914)	348 and 373
	Lugg (1968)	298
air-*i*-butyric acid	Winkelmann (1885)	371
	Pochettino (1914)	351 and 373
	Lugg (1968)	298
air-ethyl acetate	Winkelmann (1884 c)	319
	Pochettino (1914)	283 to 343
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
	Katan (1969)	295
air-methyl propionate	Winkelmann (1884 c)	319 and 340
	Griboiedov (1893)	316 and 332
	Pochettino (1914)	288 to 343
	Lugg (1968)	298
air-propyl formate	Winkelmann (1884 c)	319 and 340
	Pochettino (1914)	293 and 353
	Lugg (1968)	298
air-*p*-dioxane	Lugg (1968)	298
air-*n*-amyl alcohol	Winkelmann (1885)	372
	Lugg (1968)	298
air-active amyl alcohol	Winkelmann (1885)	372
air-*sec*-amyl alcohol	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air-ethylene glycol-monoethyl ether	Lugg (1968)	298
air-toluene	Mack (1925)	298
	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
air-*n*-butyl chloride	Grob and El-Wakil (1969)	297
air-aniline	Mack (1925)	298
	Gilliland (1934)	299 to 332
	Lugg (1968)	298
air-furfural	Brookfield et al. (1947)	298 to 323
air-fluorobenzene	Grob and El-Wakil (1969)	297
air-mesityl oxide	Lugg (1968)	298
air-$COCl_2$	Klotz and Miller (1947)	273
air-1,1-dichloroethane	Lugg (1968)	298
air-1,2-dichloroethane	Lugg (1968)	298
	Grob and El-Wakil (1969)	297
air-*n*-heptane	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*-butyl formate	Pochettino (1914)	298 to 358
	Lugg (1968)	298
air-ethyl propionate	Winkelmann (1884 c)	340 and 363
	Pochettino (1914)	283 to 366
	Fairbanks and Wilke (1950)	301
	Lugg (1968)	298
air-methyl *n*-butyrate	Winkelmann (1884 c)	340 and 365
	Pochettino (1914)	295 to 372
	Lugg (1968)	298
air-methyl *i*-butyrate	Winkelmann (1884 c)	323 and 340
	Pochettino (1914)	285 and 353
	Lugg (1968)	298
air-*n*-propyl acetate	Le Blanc and Wuppermann (1916)	315 and 340
	Pochettino (1914)	283 to 372
	Lugg (1968)	298
air-*i*-propyl acetate	Lugg (1968)	298
air-valeric acid	Pochettino (1914)	355 and 373
air-*i*-valeric acid	Winkelmann (1885)	372
	Pochettino (1914)	344 and 373
	Lugg (1968)	298
air-2-ethyl-1-butanol	Lugg (1968)	298
air-*n*-hexyl alcohol	Winkelmann (1885)	372
	Lugg (1968)	298
air-*i*-propylether	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	Pochettino (1914)	323 to 373

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

d. Other Mixtures—Continued

System	Reference	T, K
	Lugg (1968)	298
air-*m*-xylene	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air-*o*-xylene	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air-*p*-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	Altshuller 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-*n*-octane	Mack (1925)	298
	Galloway and Sage (1967)	364
	Nafikov and Usmanov (1967)	293 to 353
	Lugg (1968)	298
air-*n*-amyl formate	Pochettino (1914)	310 to 373
	Lugg (1968)	298
air-*i*-amyl formate	Pochettino (1914)	322 and 373
	Lugg (1968)	298
air-*n*-butyl acetate	Pochettino (1914)	325 and 373
	Lugg (1968)	298
air-*i*-butyl acetate	Winkelmann (1884 c)	340 and 371
	Pochettino (1914)	324 and 373
	Lugg (1968)	298
air-*n*-caproic acid	Pochettino (1914)	355 and 373
	Lugg (1968)	298
air-*i*-caproic acid	Pochettino (1914)	355 and 373
	Lugg (1968)	298
air-diacetone alcohol	Lugg (1968)	298
air-ethyl *n*-butyrate	Winkelmann (1884 c)	340 and 370
	Pochettino (1914)	315 and 373
	Lugg (1968)	298
air-ethyl *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-*n*-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-*n*-propyl benzene	Pochettino (1914)	325 to 372
	Lugg (1968)	298
air-*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-*n*-propyl bromide	Pochettino (1914)	294 and 336
	Lugg (1968)	298
air-*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-*o*-chlorotoluene	Pochettino (1914)	338 to 371
	Lugg (1968)	298
air-*m*-chlorotoluene	Pochettino (1914)	338 and 371
	Lugg (1968)	298
air-*p*-chlorotoluene	Pochettino (1914)	333 to 373
	Lugg (1968)	298
air-*n*-C_9H_{20}	Nafikov and Usmanov (1967)	293 to 353
air-napthalene	Mack (1925)	298
air-bromochloromethane	Lugg (1968)	298
air-*n*-amyl acetate	Lugg (1968)	298
air-*n*-butyl propionate	Lugg (1968)	298
air-*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* $\mathscr{D}_{12}$ *according to gas pair, temperature noted*—Continued

d. Other Mixtures—Continued

System	Reference	T, K
	Pochettino (1914)	324 to 372
	Lugg (1968)	298
air-methyl-*n*-caproate	Lugg (1968)	298
air-*n*-propyl *n*-butyrate	Winkelmann (1884 c)	371
	Pochettino (1914)	323 to 373
	Lugg (1968)	298
air-*n*-propyl *i*-butyrate	Winkelmann (1884 c)	370
	Pochettino (1914)	339 to 373
	Lugg (1968)	298
air-*i*-propyl *i*-butyrate	Pochettino (1914)	323 and 373
	Lugg (1968)	298
air-octyl alcohol	Lugg (1968)	298
air-*n*-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-*p*-cymene	Lugg (1968)	298
air-*i*-phorone	Lugg (1968)	298
air-toluene-2,4-diisocyanate	Lugg (1968)	298
air-*n*-$C_{10}H_{22}$	Altshuller and Cohen (1960)	313 to 422
	Nafikov and Usmanov (1967)	313 to 353
air-dichloroethylether	Lugg (1968)	298
air-amyl propionate	Winkelmann (1884 c)	371
	Pochettino (1914)	323 and 373
	Lugg (1968)	298
air-*i*-butyl-*n*-butyrate	Winkelmann (1884 c)	371
	Pochettino (1914)	348 to 373
	Lugg (1968)	298
air-*i*-butyl-*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-*p*-*tert*-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-*n*-$C_{11}H_{24}$	Nafikov and Usmanov (1967)	333 and 353
air-ethyl iodide	Grob and El-Wakil (1969)	297
air-amyl *n*-butyrate	Pochettino (1914)	324 and 373
	Lugg (1968)	298
air-amyl *i*-butyrate	Winkelmann (1884 c)	371
	Pochettino (1914)	357 and 373
	Lugg (1968)	298
air-*i*-butyl valerate	Winkelmann (1884 c)	371
	Pochettino (1914)	353 and 373
	Lugg (1968)	298
air-Br_2	Andrew (1955)	293
	Brockett (1966)	301
	Lugg (1968)	298
air-safrole	Pochettino (1914)	350 and 373
air-*i*-safrole	Pochettino (1914)	336 and 373
air-eugenol	Pochettino (1914)	359 and 372
air-*i*-eugenol	Pochettino (1914)	358 and 372
air-chlorpicrin	Lugg (1968)	298
air-CCl_3NO_2	Klotz and Miller (1947)	298
air-tetrachloroethylene	Lugg (1968)	298
air-1,1,2,2-tetrachlorethane	Lugg (1968)	298
air-*n*-propyl iodide	Pochettino (1914)	304 to 373
	Lugg (1968)	298
air-*i*-propyl iodide	Pochettino (1914)	324 to 352
	Lugg (1968)	298
air-*n*-$C_{12}H_{26}$	Nafikov and Usmanov (1967)	333 and 353
air-anthracene	Mack (1925)	372
air-triethyl phosphate	Lugg (1968)	298

TABLE 16. *Experimental determinations of* $\mathscr{D}_{12}$ *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 413 and 473
	Gilliland (1934)	614
	Trautz and Müller (1935)	413 and 473
	Mikhailov and Kochegarova (1967)	(?)
	Lugg (1968)	298
air-pentachloroethane	Lugg (1968)	298
air-diethyl phthalate	Lugg (1968)	298
air-*n*-$C_{16}H_{34}$	Bradley and Shellard (1949)	288 to 308
air-1,2-dibromo-3-chloropropane	Lugg (1968)	298
air-*n*-$C_{17}H_{36}$	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-*n*-$C_{18}H_{38}$	Bradley and Shellard (1949)	288 to 313
air-S_8	Bradley (1951)	303
air-tributyl phosphate	Lugg (1968)	298
air-di-*n*-butyl phthalate	Bradley et al. (1946)	293
	Birks and Bradley (1949)	288 to 313
	Lugg (1968)	298
air-tetraethyl pyrophosphate	Lugg (1968)	298
air-*bis*-2-ethylhexyl phosphate	Lugg (1968)	298
air-$(C_7H_{15})_3CH$	Bradley and Waghorn (1951)	298 to 308
air-diisooctyl phthalate	Lugg (1968)	298
air-butyl stearate	Bradley et al. (1946)	293
air-*tri*-orthocresol phosphate	Lugg (1968)	298
air-$(C_{10}H_{21})_3CH$	Bradley and Waghorn (1951)	298 to 308
air-*n*-$C_{16}F_{34}$	Bradley and Waghorn (1951)	288 to 303
CH_3OH-CO_2	Winkelmann (1885)	299 and 323
CH_3OH-1-butanol	Weissmann (1968 a)	423
H_2S-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_3H_6-C_3H_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_2O	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_3H_8	Wall and Kidder (1946)	298
	Weissman (1964)	300 to 550
CO_2-formic acid	Winkelmann (1885)	339 and 358
CO_2-C_2H_5OH	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-*n*-propyl alcohol	Winkelmann (1885)	340 and 357
CO_2-*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* $\mathscr{D}_{12}$ *according to gas pair, temperature noted*—Continued

d. Other Mixtures—Continued

System	Reference	*T*, K
CO_2-*n*-butyl alcohol	Winkelmann (1885)	372
CO_2-*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-*n*-butyric acid	Winkelmann (1885)	372
CO_2-*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-*n*-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*-butyrate	Winkelmann (1884 c)	323 and 340
CO_2-*i*-valeric acid	Winkelmann (1885)	372
CO_2-*n*-hexyl alcohol	Winkelmann (1885)	372
CO_2-*i*-butyl acetate	Winkelmann (1884 c)	340 and 371
CO_2-ethyl butyrate	Winkelmann (1884 c)	340 and 370
CO_2-ethyl *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*-butyl propionate	Winkelmann (1884 c)	371
CO_2-propyl butyrate	Winkelmann (1884 c)	371
CO_2-propyl-*i*-butyrate	Winkelmann (1884 c)	370
CO_2-amyl propionate	Winkelmann (1884 c)	371
CO_2-*i*-butyl butyrate	Winkelmann (1884 c)	371
CO_2-*i*-butyl-*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*-butyrate	Winkelmann (1884 c)	371
CO_2-*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_2O-ethylene oxide	Wall and Kidder (1946)	298
N_2O-C_3H_8	Wall and Kidder (1946)	298
	Weissman (1964)	300 to 550
N_2O-Br_2	Mackenzie and Melville (1933)	290
C_3H_8-*n*-hexane	Carmichael et al. (1955 b)	294 to 377
NO_2-N_2O_4	Weissman (1968 a)	303 to 343
C_2H_5OH-C_4H_9OH	Weissman (1968 a)	423
C_2H_5OH-CCl_2F_2	Lee and Wilke (1954)	298
$(CH_3)_2O$-CH_3Cl	Chakraborti and Gray (1966)	303 to 333
	Weissman (1968 a)	308 and 353
$(CH_3)_2O$-SO_2	Chakraborti and Gray (1966)	303 to 333
	Weissman (1968 a)	308 and 353
CH_3Cl-SO_2	Chakraborti and Gray (1966)	303 to 333
	Weissman (1968 a)	308 and 353
CH_3Cl-C_2H_5Cl	Manner (1967)	298 to 419
1,3-butadiene-1-butyne	Bournia et al. (1961)	300
C_3H_7OH-C_4H_9OH	Weissman (1968 a)	423
SO_2-SO_2F_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
n-C_5H_{12}-*n*-C_5H_{12}	Beatty (1969)	273
n-C_5H_{12}-$C(CH_3)_4$	Beatty (1969)	273
$(C_2H_5)_2O$-$CHCl_3$	Weissman (1964)	293
C_6H_6-CCl_4	Weissman (1964)	293
C_6H_6-CCl_2F_2	Lee and Wilke (1954)	298
HBr-DBr	Braune and Zehle (1941)	294—296
cyclohexane-Rb	McNeal (1962)	323
	Violino (1968)	323
CH_2Cl_2-CCl_4	Weissman (1964)	293 to 413
CF_4-CF_4	Khoury and Kobayashi (1970)	243 to 348
CF_4-SF_6	Raw and Tang (1963)	303 to 342
i-octane-$C_6H_5NO_2$	Huber and van Vught (1965)	298
triethylamine-CCl_2F_2	Mehta (1966)	298
C_8H_{18}-C_7F_{16}	Weissman (1964)	303 and 323
BCl_3-CCl_4	Raw (1955)	303
$CHCl_3$-CCl_4	Weissman (1964)	293
Cl_2CF_2-Cl_2CF_2	Miller and Carman (1961)	293

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

d. Other Mixtures—Continued

System	Reference	*T*, K
CCl_2F_2-di-*n*-butyl phthalate	Birks and Bradley (1949)	293 and 303
Hg-Hg	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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Byrne, J. J., Maguire, D., and Clarke, J. K. A. (1967), J. Phys. Chem. **71,** 3051; nitromethane-(Ar, H_2, N_2), monofluorobenzene-(Ar, H_2, N_2), 1-bromo-3-methylbutane-(Ar, H_2, N_2), 1-iodopropane-(H_2, N_2), 1-bromobutane-(H_2, N_2), evaporation tube.

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Chang, K. C., Hesse, R. J., and Raw, C. J. G. (1970), Trans. Faraday Soc. **66,** 590; SO_2-SO_2F_2, mixture viscosity.

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Cummings, G. A. McD., and Ubbelohde, A. R. (1955), J. Chem. Soc., p. 2524; *n*-octane-(H_2, N_2), N_2-*n*-decane, 2,2,4-trimethylpentane-(H_2, N_2), and recalculations for preceding reference, evaporation tube.

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DiPippo, R., Kestin, J., and Oguchi, K. (1967), J. Chem. Phys. **46,** 4986; He-(Ne, Ar, CO_2), Ne-(Ar, N_2), Ar-(NH_3, N_2), mixture viscosity.

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DuBro, G. A., and Weissman, S. (1970), Phys. Fluids **13,** 2682; ^{3}He-^{4}He, He-(Ne, Ar), two-bulb apparatus.

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Ember, G., Ferron, J. R., and Wohl, K. (1964), AIChE J. **10,** 68; CH_4-CH_4, CO_2-(CH_4, H_2O, CO_2), point source.

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Fejes, P., and Czárán, L. (1961), Hungary Acta Chim. **29,** 171; H_2-(CH_4, N_2, C_2H_6, C_3H_8, butane), CH_4-CO_2, N_2-(C_2H_6, C_3H_8, butane), gas chromatography.

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Ferron, J. R., and Dunham, P. G. (1967), Ind. Chim. Belge **32,** (Special number), part 1, 313; He-CO_2, shocktube.

Foch, R. (1913), Ann. Chim. Phys. **29,** 597; air-CO_2, open tube.

Franzen, W. (1959), Phys. Rev. **115,** 850; Rb-(Ne, Ar, Kr, Xe), optical pumping.

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Fuller, E. N., and Giddings, J. C. (1967), in Giddings and Mallik (1967); N_2-(C_2H_4, butane), gas chromatography.

Fuller, E. N., Ensley, K., and Giddings, J. C. (1969), J. Phys. Chem. **73,** 3679; N_2-(C_2H_4, butane), He-(1-fluorohexane, fluorobenzene, difluoromethane, 1,1-difluorethane, hexafluorobenzene, 4-fluorotoluene, dichloromethane, trichloromethane, 1,2-dichloroethane, 1-chloropropane, 1-chlorobutane, 2-chlorobutane, 1-chloropentane, chlorobenzene, dibromomethane, bromoethane, 1-bromopropane, 2-bromopropane, 1-bromobutane, 2-bromobutane, 1-bromohexane, 2-bromohexane, 3-bromohexane, bromobenzene, 2-bromo-1-chloropropane, iodomethane, iodoethane, 1-iodopropane, 2-iodopropane, 1-iodobutane, 2-iodobutane), gas chromatography.

Gaede, W. (1915), Ann. Physik **46,** 357; Hg-(H_2, air), condensable vapor pumping effect and evaporation tube.

Galloway, T. R., and Sage, B. H. (1967); Chem. Eng. Sci. **22,** 979; air-(*n*-hexane, *n*-heptane, *n*-octane), evaporation tube.

Getzinger, R. W., and Wilke, C. R. (1967), AIChE J. **13,** 577; air-(CH_3OH, ethanol, benzene, $CHCl_3$, CCl_4), evaporation tube.

Giddings, J. C. (1968), Private communication; H_2-He, gas chromatography.

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Giddings, J. C., and Seager, S. L. (1962), Ind. Eng. Chem., Fundam. **1,** 277; He-(Ar, NH_3, N_2, O_2, CO_2), H_2-(He, N_2, CO_2), N_2-(O_2, CO_2), gas chromatography.

Gilliland, E. R. (1934), Ind. Eng. Chem. **26,** 681; air-(H_2O, 2-propanol, 1-butanol, 2-butanol, 2-pentanol, toluene, ethyl acetate, chlorobenzene, aniline, diphenyl, Hg), evaporation tube.

Ginsel, L. A., and Ornstein, L. S. (1933), Z. Physik **84,** 276; Na-flame atmosphere, flame diffusion.

Golubev, I. F., and Bondarenko, A. G. (1963), Gazov. Prom. **8,** 46; Ar-(He, H_2), two-bulb apparatus.

Goryunova, N. A., and Kuvshinskiĭ, E. V. (1948), Zh. Tekh. Fiz. (USSR) **18,** 1421; air-(acetone, $CHCl_3$, cyclohexane), evaporation tube.

Gover, T. A. (1967), J. Chem. Educ. **44,** 409; C_2H_6-(CH_4, C_3H_8, CO_2), closed tube.

Gozzini, A., Ioli, N., and Strumia, F. (1967), Nuovo Cimento **B49,** 185; He-Na, optical pumping.

Griboiedov, S. (1893), J. Russ. Phys-Chem. Soc. **25,** 36; air-(benzene, ethyl ether, methyl formate, methyl acetate, methyl propionate), evaporation tube.

Grieveson, P., and Turkdogan, E. T. (1964), J. Phys. Chem. **68,** 1547; Ar-(Cr, Fe, Co, Ni), evaporation tube.

Grob, A. K., and El-Wakil, M. M. (1969), Trans. ASME, J. Heat Transfer **91C,** 259; air-(*n*-hexane, *n*-heptane, benzene, CCl_4, ethyl acetate, 1,2-dichloroethane, ethyl iodide, fluorobenzene, *n*-butyl chloride, ethyl bromide), unsteady evaporation.

Groth, W., and Harteck, P. (1941), Z. Elektrochem. **47,** 167; Kr-Kr, Xe-Xe, closed tube.

Groth, W., and Sussner, E. (1944), Z. Physik. Chem. (Leipzig) **A193,** 296; Ne-Ne, closed tube.

Guglielmo, G. (1881), Atti Accad. Torino **17,** 54; H_2O-air, evaporation tube.

Guglielmo, G. (1882), Atti Accad. Torino **18,** 93; H_2O-(H_2, air, CO_2), evaporation tube.

Gurvich, V. S., and Matizen, E. V. (1968), Isv. Sib. Otd. Akad. Nauk SSSR, Ser. Khim. Nauk **6,** 8; in Chem. Abstr. **70,** 71226 j (1969); Ar-CO_2, capillary method (?); method not given in abstract.

Gush, L. L. (1948), Trans. Inst. Chem. Eng. (London) **26,** 142; air-acetone, evaporation tube.

Hargrove, G. L., and Sawyer, D. T. (1967), Anal. Chem. **39,** 244; He-(ethanol, *n*-butane, *n*-pentane, *n*-hexane, ether, acetone, benzene), Ar-(*n*-butane, *n*-pentane, *n*-hexane, ether, acetone, benzene), N_2-*n*-butane, gas chromatography.

Harteck, P., and Schmidt, H. W. (1933), Z. Physik. Chem. (Leipzig) **B21,** 447; *p*-H_2 in normal-H_2, closed tube and back diffusion.

Hartland, A., and Lipsicas, M. (1963), Phys. Letters **3,** 212; H_2-H_2, nuclear magnetic resonance.

Hawtin, P., Dawson, R. W., and Roberts, J. (1969), Trans. Inst. Chem. Eng. (London) **47,** T109; He-(Ar, N_2), CH_4-CO_2, diffusion bridge (porous septum).

Heath, H. R., Ibbs, T. L., and Wild, N. E. (1941), Proc. Royal Soc. **A178,** 380; H_2-D_2, closed tube.

Heinzelmann, F. J., Wasan, D. T., and Wilke, C. R. (1965), Ind. Eng. Chem., Fundam. **4,** 55; air-benzene, evaporation tube.

Henry, J. P., Jr., Cunningham, R. S., and Geankoplis, C. J. (1967), Chem. Eng. Sci. **22,** 11; He-N_2, diffusion bridge (porous septum).

Hippenmeyer, B. (1949), Z. Angew. Phys. **1,** 549; H_2O-(H_2, N_2), evaporation tube.

Hirst, W., and Harrison, C. E. (1939), Proc. Royal Soc. **A169,** 573; Rn-(He, Ne, Ar, H_2, air), closed tube.

Hogervorst, W. (1971), Physica **51,** 59; He-(Ne, Ar, Kr, Xe), Ne-(Ar, Kr, Xe), Ar-(Kr, Xe), cataphoresis.

Hogervorst, W., and Freudenthal, J. (1967), Physica **37,** 97; Ne-Ar, cataphoresis.

Holmes, R., and Tempest, W. (1960), Proc. Phys. Soc. (London) **75,** 898; He-(Ne, Ar, Kr), sound absorption.

Holsen, J. N., and Strunk, M. R. (1964), Ind. Eng. Chem., Fundam. **3,** 143; He-(Ar, air, CO_2), CO_2-(Ar, air), closed tube.

Houdaille (1896), Thèses, Paris; in Landolt-Börnstein, Physikalisch-Chemische Tabellen (J. Springer, Berlin, 1923) Vol. **I,** p. 251; H_2O-air, evaporation tube.

Houghton, H. G. (1933), J. Appl. Phys. **4,** 419; H_2O-air (at various humidities), droplet evaporation.

Hu, A. T-C., and Kobayashi, R. (1970), J. Chem. Eng. Data **15,** 328; He-(Ar, CH_4, N_2, CO_2), CH_4-(CH_4, CH_3T, CF_4), CH_3T-CF_4, gas chromatography.

Huang, T-C., Sheng, S-J., and Yang, F. J. F. (1968), J. Chin. Chem. Soc. (Taipei) **15,** 127, H_2-(benzene, toluene, *n*-pentane, *n*-hexane, cyclohexane, ethanol, methanol, butanol-1, butanol-2), gas chromatography.

Huber, J. F. K., and van Vught, G. (1965), Ber. Bunsenges. Phys. Chem. **69,** 821; N_2-*n*-hexane, nitrobenzene-*i*-octane, gas chromatography.

Hudson, G. H., McCoubrey, J. C., and Ubbelohde, A. R. (1960), Trans. Faraday Soc. **56,** 1144; benzene-(H_2, N_2, O_2), cyclohexane-(H_2, N_2, O_2), pyridine-(H_2, N_2, O_2), piperidine-(H_2, N_2, O_2), thiophen-(H_2, N_2, O_2), tetrahydrothiophen-(H_2, N_2, O_2), evaporation tube.

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Humphreys, A. E., and Mason, E. A. (1970), Phys. Fluids **13,** 65; Ar-Kr, two-bulb apparatus and temperature dependence of thermal diffusion factor.

Hutchinson, F. (1947), Phys. Rev. **72,** 1256; Ar-Ar, closed tube.

Hutchinson, F. (1949), J. Chem. Phys. **17,** 1081; Ar-Ar, two-bulb apparatus.

Islam, M., and Stryland, J. C. (1969), Physica **45,** 115 (reports result by A. I. Carswell, Ph. D. Thesis, Univ. Toronto, 1960); Ar-CH_4, closed tube.

Ivakin, B. A., and Suetin, P. E. (1964 a), Soviet Phys.-Tech. Phys. **8,** 748 [Zh. Tekh. Fiz. **33,** 1007 (1963)]; H_2-(D_2, NH_3, CO), D_2-(He, Ar, NH_3, N_2, CO, air, CO_2, SF_6), NH_3-(He, Ar, N_2, CO, air, SF_6), CO-(He, Ar, N_2, CO_2, SF_6), closed tube.

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Ivanovskiĭ, M. N., Sorokin, V. P. Subbotin, V. I., and Chulkov, B. A. (1969), High Temp. (USSR) **7,** 433 [Teplofiz. Vys. Temp. **7,** 479 (1969)]; K-(He, Ar, H_2, N_2), diffusion-controlled condensation.

Jacobs, T., Peeters, L., and Vermant, J. (1970), Bull. Soc. Chim. Belges **79,** 337; Ar-(CH_4, C_2H_6, C_3H_8, *n*-C_4H_{10}), N_2-(CH_4, C_2H_6, C_3H_8, *n*-C_4H_{10}), closed tube.

Jona, F. (1965), J. Chem. Phys. **42,** 1025; $GeBr_2$-$GeBr_4$, GeI_2-GeI_4, diffusion-controlled evaporation.

Jorgensen, F., and Watts, H. (1961), Chem. Ind. (London), p. 1440; air-benzene, evaporation tube.

Kalelkar, A. S., and Kestin, J. (1970), J. Chem. Phys. **52,** 4248; He-(Ar, Kr), mixture viscosity.

Kamnev, A. B., and Leonas, V. B. (1966), High Temp. (USSR) **4,** 283 [Teplofiz. Vys. Temp. **4,** 288 (1966)]; Kr-Kr, Xe-Xe, molecular beam scattering.

Katan, T. (1969), J. Chem. Phys. **50,** 233; air-(methanol, ethanol, benzene, ethyl acetate), droplet evaporation (modified).

Kaufmann (1967), in Frost (1967); He-(C_2H_6, C_3H_8); open tube.

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Kestin, J., Kobayashi, Y., and Wood, R. T. (1966), Physica **32,** 1065; He-(Kr, N_2), CO_2-(Ar, N_2), mixture viscosity.

Kestin, J., Wakeham, W., and Watanabe, K. (1970), J. Chem. Phys. **53,** 3773; Ar-(Ne, Kr), mixture viscosity.

Khomchenkov, B. M., Aref'ev, K. M., Borishanskiĭ, V. M., Paleev, I. I., Ivashchenko, N. I., Ulitskiĭ, R. I., Kholmiskiĭ, I. G., and Suslova, L. A. (1968), High Temp. (USSR) **6,** 956 [Teplofiz. Vys. Temp. **6,** 999 (1968)]; He-(K, Cs), Ar-(K, Cs), evaporation tube.

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Mian, A. A. (1967), Ph.D. Thesis, Louisiana State Univ., Baton Rouge, Ar-(He, HBr), N_2-(He, HCl, CO_2, HBr), diffusion bridge (porous septum).

Mian, A. A., Coates, J., and Cordiner, J. B. (1969), Canadian J. Chem. Eng. **47**, 499; Ar-HBr, N_2-(HCl, HBr), diffusion bridge (porous septum).

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Naccari, A. (1910), Nuovo Cimento **19**, 52; air-ether, evaporation tube.

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Rutherford, E., and Brooks, H. T. (1901), Trans. Royal Soc. Canada **7**, 21; Rn-air, closed tube.

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Saran, A., and Singh, Y. (1966), Canadian J. Chem. **44**, 2222; Kr-(Kr, SO_2), two-bulb apparatus.

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*A misprint appears in table 1; the highest temperature for CH_4-H_2O is not 323 K but 328 K.

O_2, air, CO_2, SF_6), Ar-(C_2H_2, CO_2, SF_6), H_2-(He, Ar, N_2, air, CO_2, SF_6), N_2-(CO_2, SF_6), O_2-(C_2H_2, CO_2, SF_6), air-(CO_2, SF_6), N_2-(CO_2, SF_6), O_2-(acetylene, CO_2, SF_6), air-(CO_2, acetylene, SF_6), CO_2-SF_6, closed tube.

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The gas pairs investigated and the potential energy separation ranges are noted.

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Belyaev, Yu. N., and Leonas, V. B. (1966 a), Soviet Phys.-Tech. Phys. **11,** 257 [Zh. Tekh. Fiz. **36,** 353 (1966)]; Ar-N_2(2.26–2.86 Å), Ar-O_2(2.18–2.94 Å), N_2-N_2(2.34–3.05 Å), N_2-O_2(2.34–3.05 Å), O_2-O_2(2.34–3.15 Å). Errata in Soviet Phys.-Doklady **11,** 866 (1967).

Belyaev, Yu. N., and Leonas, V. B. (1966 b), Soviet Phys.-JETP Letters **4,** 92 [Zh. Eksper. Teor. Fiz. Pis'ma **4,** 134 (1966)]; H-N_2(1.46–1.81 Å), H-O_2(1.46–1.84 Å), N-N_2(1.76–2.54 Å), N-O_2(1.84–2.49 Å), O-N_2(2.00–2.48 Å), O-O_2 (2.05–2.46 Å).

Belyaev, Yu. N., and Leonas, V. B. (1966 c), High Temp. (USSR) **4,** 686 [Teplofiz. Vys. Temp **4,** 732 (1966)]; same results as reported by Belyaev and Leonas (1966 a).

Belyaev, Yu. N., and Leonas, V. B. (1967 a), Soviet Phys.-Doklady **11,** 866 [Dokl. Akad. Nauk SSSR **170,** 1039 (1966)]; Ar-N_2 (2.12–2.67 Å), Ar-O_2(2.15–2.63 Å), N_2-N_2(2.34–3.05 Å), N_2-O_2 (2.34–3.05 Å), O_2-O_2(2.34–3.15 Å).

Belyaev, Yu. N., and Leonas, V. B. (1967 b), Soviet Phys.-Doklady **12,** 233 [Dokl. Akad. Nauk SSSR **173,** 306 (1967)]; He-He (1.10–1.53 Å), He-H_2(1.15–1.89 Å), H_2-H_2(1.34–1.95 Å), H-He (0.79–1.35 Å), H-H_2(1.00–1.24 Å).

Belyaev, Yu. N., and Leonas, V. B. (1967 c), High Temp. (USSR) **5,** 1014 [Teplofiz. Vys. Temp. **5,** 1123 (1967)]; same results as reported by Belyaev and Leonas (1967 b).

Belyaev, Yu. N., and Leonas, V. B. (1968), High Temp. (USSR) **6,** 182 [Teplofiz. Vys. Temp. **6,** 188 (1968)]; N_2-N_2(2.18–2.72 Å), N_2-O_2(2.08–2.68 Å), O_2-O_2(2.05–2.75 Å); other results as reported by Belyaev and Leonas (1966 b).

Belyaev, Yu. N. Leonas, V. B., and Sermyaguin, A. V. (1967), Private communication from V. B. Leonas; in Abstracts of Papers, Fifth International Conference on the Physics of Electronic and Atomic Collisions (Publishing House "Nauka," Leningrad, USSR, 1967), p. 643; N_2-NO(2.28–2.85 Å), N_2-CO(2.22–2.77 Å), CO-CO(2.31–2.92 Å), N-NO(1.90–2.40 Å), O-Ar(1.78–2.40 Å), O-Kr(1.91–2.24 Å), O-Xe(2.08–2.37 Å), F-Ar(1.80–2.22 Å), F-Kr(1.93–2.28 Å), F-Xe(2.03–2.33 Å).

Belyaev, Yu. N., Kamyshov, N. V., Leonas, V. B., and Sermyagin, A. V. (1969), Entropie **30,** 173; CH_4-CH_4(2.14–2.62 Å), NH_3-NH_3(1.51–2.23 Å), N_2-CO(2.10–2.62 Å), N_2-NO(2.05–2.55 Å), N_2-CO_2(2.60–3.16 Å), CO-CO(2.10–2.65 Å), CO-NO(2.05–2.65 Å), CO-O_2(2.00–2.60 Å), CO-CO_2(2.47–3.01 Å), NO-NO(2.00–2.64 Å), NO-O_2(2.04–2.58 Å), O_2-CO_2(2.27–2.81 Å), CO_2-CO_2 (2.73–3.26 Å), N-CH_4(1.83–2.38 Å), O-CO_2(2.08–2.60 Å). These results appeared too late for inclusion in the correlation.

Kamnev, A. B., and Leonas, V. B. (1965 a), Soviet Phys.-Doklady **10,** 529 [Dokl. Akad. Nauk SSSR **162,** 798 (1965)]; He-He (0.87–1.27 Å), He-Ne(1.3–1.65 Å), He-Ar(1.63–2.06 Å), Ne-Ne (1.7–2.18 Å), Ne-Ar(1.93–2.49 Å), Ar-Ar(2.26–3.14 Å).

Kamnev, A. B., and Leonas, V. B. (1965 b), High Temp. (USSR) **3,** 744 [Teplofiz. Vys. Temp. **3,** 804 (1965)]; same results as reported by Kamnev and Leonas (1965 a).

Kamnev, A. B., and Leonas, V. B. (1966 a), Soviet Phys.-Doklady **10**, 1202 [Dokl. Akad. Nauk SSSR **165**, 1273 (1965)]; He-Kr (1.67–2.04 Å), He-Xe(1.73–2.2 Å), Ne-Kr(2.15–2.52 Å), Ne-Xe (2–2.56 Å), Ar-Kr(2.4–3.1 Å), Ar-Xe(2.48–3.27 Å), Kr-Kr(2.4–3.02 Å), Kr-Xe(2.44–3 Å), Xe-Xe(2.48–3.09 Å).

Kamnev, A. B., and Leonas, V. B. (1966 b), High Temp. (USSR) **4**, 283 [Teplofiz. Vys. Temp. **4**, 288 (1966)]; same results as reported by Kamnev and Leonas (1966 a).

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