error of 0.05 mV. For the HCl–*t*B·HCl system, the deviation is 0.0008 in log γ_{HCl}. These deviations are trivial from the point of view of a calculation of activity coefficients. However, the data are fitted significantly better by the complete Equation 2, as shown by the $\Sigma\Delta^2$ figures given in Table III, which demonstrates that the deviations from linearity are systematic rather than random. We cautiously conclude, therefore, that deviations from the simple Harned relation exist at an ionic strength of 1.0 mol kg^{-1}, in contrast with the normal situation exemplified by the results for the HCl–NH_4Cl system.

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Binary Gaseous Diffusion Coefficients
IV. Sulfur Hexafluoride with *n*-Hexane, *n*-Heptane, *n*-Octane, and 2,2,4-Trimethylpentane at 1 Atm at 10–70°C

Emmerich Wilhelm
Institut für Physikalische Chemie, Universität Wien, Währingerstrasse 42, A-1090 Wien, Austria

Rubin Battino[1]
Department of Chemistry, Wright State University, Dayton, Ohio 45431

Binary gaseous diffusion coefficients at 1-atm pressure of sulfur hexafluoride, with *n*-hexane, *n*-heptane, *n*-octane, and 2,2,4-trimethylpentane (isooctane) were measured at 10°, 25°, 40°, 55°, and 70°C by the capillary tube method of Stefan. Diffusion coefficients were calculated (a) by using Lennard-Jones (6, 12) pair potential parameters in conjunction with the Hudson-McCoubrey combining rule and (b) with a semiempirical method somewhat similar to that suggested by Chen and Othmer. Diffusion coefficients calculated via method (b) were in much better agreement with experiment than those obtained via method (a). The experimental results were used to investigate certain regularities in diffusional behavior of homologous species in mixtures with a common gas. In particular, a strikingly simple correlation (first advanced by Wakeham and Slater) was tested and found applicable, even when the common gaseous component is a pseudospherical molecule of considerable size and mass, such as SF_6. Provided that experimental data on two sufficiently spaced homologs are available, the correlation apparently offers a powerful predictive potential.

Several aspects of the diffusional behavior of polyatomic gases in mixtures with a common gas have recently been investigated in great detail (*1, 6–10, 16, 23, 28, 34, 35*). In particular, a simple and hitherto unexpected regularity within a homologous series (such as *n*-alkanes) for the binary diffusion coefficient D_{12} of members of such a series in a common gas has been reported by Elliott and Watts (*6, 7*). This heuristic correlation connected the experimental diffusion coefficients with the reduced mass $\mu = M_1M_2/(M_1 + M_2)$ of the relevant species, that is

$$D_{12} = A\mu + B \qquad (1)$$

Here, A and B are temperature dependent constants with $-B/A$ independent of temperature and approximately equal to the molar mass M_1 of the common partner gas; M_2 represents the molar mass of the homolog. However, an elementary analysis by Humphreys and Mills (*13*), with the aid of a simple rigid sphere model, revealed that linear behavior is not to be expected over extreme ranges of μ: theory predicts slightly S-shaped curves. Nevertheless, for limited ranges of μ, little deviation from linearity is predicted, and the theoretical curves meet the μ-axis at $\mu = M_1$ as indeed they must, since $\lim_{M_2 \to \infty} D_{12} = 0$. In fact, detailed analysis of the then available data indicated that deviations from linearity do exist, both in the series *n*-alkanes–air (*6*) and *n*-alkanes–nitrogen (*14*), although the experimental basis for drawing definite conclusions was rather small. Thus, to discern deviations from simple linear behavior, it was suggested that experiments be carried out either with higher members of homologous series (i.e., to cover the range $\mu/M_1 \to 1$) or with a more massive common gas (i.e., to investigate the region of small μ/M_1).

Following this suggestion, Wakeham and Slater (*29*) measured diffusion coefficients at 1 atm for the series methane through butane in argon, and combining their results with literature data for higher *n*-alkanes, were able to show conclusively that deviations from a linear plot D_{12} vs. μ exist (cf. also ref. *28*). Prior to this work we have studied the diffusional behavior of members of the *n*-alkane series with the nonpolar

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[1] To whom correspondence should be addressed.

pseudosphericals methane and carbon tetrafluoride (*34*). That is, we studied the variation of D_{12} with increasing size (and mass) of the common gas. The results reported in this publication are an extension of our earlier work (*34*) to include sulfur hexafluoride. Thus, a considerable body of experimental data on D_{12} of alkanes with gases ranging in relative molecular mass up to 146 (SF_6) is now available.

Experimental

The experimental procedure was identical to that described earlier (*1, 34, 35*) and quite similar to the modified Stefan (*27*) apparatus reported by Kohn and Romero (*16*). Four diffusion tubes made of 2-mm i.d. precision bore tubing and 20 cm long with ground tapered ends (for easy sighting) were used to run four solvents and one gas at one time. All readings of diffusion path lengths were made in triplicate to 0.01 mm, but the reproducibility was about 0.05 mm. The 20-gal water bath was controlled to ±0.002°C with a Tronac thermoregulator, and temperatures were determined to ±0.002°C with a calibrated platinum resistance thermometer.

Pressure in the system was controlled to ±2 mm Hg by use of a mercury-filled cartesian manostat. The manometer was read to 0.2 mm, and the barometer (all corrections applied) to 0.1 mm. Gas flow rates were determined with a stop watch and a soap bubble flow meter. Flow rates ranged from 30 to 130 ml/min. Times were determined on a synchronous digital timer to ±0.1 min and checked out at this accuracy when compared to NBS station WWV. Liquids were saturated with the gas before transfer to the diffusion tubes, and the saturated liquids were equilibrated for 1–2 hr to assure no outgassing during a run. Runs took 2–8 days. No end effects were detected using widely varying diffusion path lengths.

A check at 25°C for the system H_2O–N_2 yielded a value of $D_{12} = 0.250$ cm^2 sec^{-1} which has to be compared with a "best" literature value (*19*) of 0.251 cm^2 sec^{-1}. The agreement is quite satisfactory since Mason and Marrero (*19, 20*) consider the very best diffusion work to be imprecise to ±1%.

The solvents used were all Phillips pure grade (99 mol % minimum purity) and were used straight from the bottle. Sulfur hexafluoride was the purest grade available from Matheson Gas Products, Inc. Its composition was greater than 99 mol % of the major constituent, and the gas was used without further purification.

Results

Experimental results are given in Table I for approximately 10°, 25°, 40°, 55°, and 70°C and 1-atm (101.325 kPa) pressure. The binary diffusion coefficients were calculated (*24*) from Equation 1:

$$D_{12} = \frac{L_\theta^2 - L_0^2}{2\,\theta}\,\frac{RT\rho_L}{PM_L}\,\frac{1}{2.303 \log\left[P/(P - P_L)\right]} \qquad (2)$$

Here, L_0 and L_θ are the diffusion paths at time zero and θ, respectively, R is the gas constant, T is the thermodynamic temperature, P is the total experimental pressure of the system, P_L is the saturation vapor presure of the liquid at temperature T, ρ_L is the liquid density, and M_L is the molar mass of the liquid. Densities and vapor pressures were obtained from standard references (*25, 36*).

According to Equation 2, the diffusion coefficient may be calculated from the slope of a plot ($L_\theta^2 - L_0^2$) vs. θ. As usual, we obtained D_{12} by determining the average of the slopes for each datum point at time θ referred back to time zero. Thus, each point is treated as a discrete experiment. The average deviation of the slopes determined in this manner was of the order of ±1%, the high-temperature values being more precise than the low-temperature values.

Conversion to a pressure of P_1 = 1 atm (101.325 kPa) was performed by making use of the fact that for moderate pressures the magnitude of the diffusion coefficient is inversely proportional to the pressure (*26, 37*):

$$(D_{12})_P/(D_{12})_{P_1} = P_1/P \qquad (3)$$

The correction did not exceed 2% for any case. The resulting values of D_{12} at various temperatures are given in Table I.

The temperature dependence of the diffusion coefficient for each system was well reproduced by fitting the experimental data to the logarithmic form of the equation $(D_{12})_T = (D_{12})_{T_\text{ref}}\,(T/T_\text{ref})^n$, where $(D_{12})_T$ is the diffusion coefficient at temperature T and 1 atm, and $(D_{12})_{T_\text{ref}}$ is the diffusion coefficient at the same pressure and a reference temperature T_ref, conveniently taken as 1K.

$$\log\,(D_{12}/\text{cm}^2\,\text{sec}^{-1})_T = \log\,(D_{12}/\text{cm}^2\,\text{sec}^{-1})_{1\text{K}} + n \log\,(T/\text{K}) \qquad (4)$$

The constant n is, in general, a nonintegral exponent, characteristic for a particular pair. Table II summarizes the value of n and $(D_{12})_{1\text{K}}$ for the systems in Table I.

The average deviations of the experimental diffusion coefficients from those calculated using Equation 4 are also tabulated in Table II; they are of the order of ±1%. Deviation plots showed no systematic bias. It is interesting to note that, with the exception of *n*-hexane/SF_4, for all sulfur hexafluoride systems a substantially larger value for the exponent n is obtained than with comparable CF_4 systems (*34*).

Discussion

It is convenient to discuss the experimental results under two separate sections.

Table I. Experimental and Calculated Binary Gaseous Diffusion Coefficients D_{12} at 1-Atm Pressure

	D_{12}, cm^2 sec^{-1}			D_{12}, cm^2 sec^{-1}		
t, °C	Exptl	Calcd, method (a)	Calcd, method (b)	Exptl	Calcd, method (a)	Calcd, method (b)
		SF_6–*n*-hexane			SF_6–*n*-heptane	
10.00	0.0285	0.0240	0.0264	0.0242	0.0217	0.0231
25.03	0.0316	0.0266	0.0292	0.0275	0.0241	0.0257
40.01	0.0351	0.0293	0.0322	0.0311	0.0266	0.0283
55.00	0.0380	0.0322	0.0353	0.0344	0.0292	0.0310
69.96	...	...	...	0.0374	0.0319	0.0339
		SF_6–*n*-octane			SF_6–2,2,4-trimethylpentane	
10.00	0.0219	0.0197	0.0207	...	0.0199	0.0214
25.03	0.0244	0.0219	0.0230	0.0253	0.0221	0.0238
40.01	0.0277	0.0241	0.0253	0.0285	0.0243	0.0262
55.00	0.0311	0.0265	0.0278	0.0308	0.0267	0.0287
69.96	0.0336	0.0289	0.0303	0.0336	0.0292	0.0314

Table II. Parameters in Equation 4 for Diffusion of Stated Component in SF_6 in Temperature Range 10–70 °C and at 1-Atm Pressure

Component	$10^6[D_{12}]_{1\text{K}}$, cm^2 sec^{-1}	n	Rms dev, cm^2 sec^{-1}
n-Hexane	0.4236	1.969	0.0002
n-Heptane	0.0594	2.289	0.0004
n-Octane	0.0551	2.283	0.0004
2,2,4-Trimethylpentane	0.3007	1.992	0.0003

Prediction of diffusion coefficients with aid of semiempirical potential parameters. At low pressures the kinetic theory of gases (*11*) provides the following expression for the binary diffusion coefficient (first approximation):

$$D_{12} = \frac{0.0018583\ T^{3/2}}{P\sigma_{12}^2 \mu^{1/2}\ \Omega_{12}^{(1,1)*}(T_{12}^*)} \tag{5}$$

The details of the collision dynamics are represented by the collision integral $\Omega_{12}^{(1,1)*}(T_{12}^*)$ at a reduced temperature $T_{12}^* = kT/\epsilon_{12}$. The molecular potential energy parameters characteristic of a 1-2 interaction are denoted by σ_{12} and ϵ_{12}, respectively. Two methods are used in computing these mixed parameters:

Method a. Lennard-Jones (6, 12) pair potential parameters for the pure components with subscripts 1 and 2 (obtained from refs. *11, 24,* and *33*) were used in conjunction with the Hudson-McCoubrey combining rule (*12*), a procedure extensively used by Watts (*30, 31*):

$$\sigma_{12} = (\sigma_1 + \sigma_2)/2 \tag{6}$$

and

$$\epsilon_{12} = (\epsilon_1\epsilon_2)^{1/2}\ \frac{2(I_1I_2)^{1/2}}{I_1 + I_2}\ \frac{\sigma_1{}^3\sigma_2{}^3}{\sigma_{12}^6} \tag{7}$$

Here, I denotes the ionization potentials, which were taken from Landolt-Börnstein (*18*); the value of I for 2,2,4-trimethylpentane was estimated by using the regular behavior of this quantity as in the case of hexane isomers. Pertinent data are summarized in Table III.

Method b. A quite successful method for evaluating potential parameters was introduced by Chen and Othmer (*3*), who give the following expressions for the pure components:

$$\epsilon/k = 1.276\,T_c^{0.9061} \tag{8}$$

$$\sigma = 0.5894\,V_c^{0.4006} \tag{9}$$

Here, T_c and V_c denote the critical temperature and volume, respectively (see Table III). For the mixed parameters, these authors suggest the simple conventional combining rules $\sigma_{12} = (\sigma_1 + \sigma_2)/2$ and $\epsilon_{12} = (\epsilon_1\epsilon_2)^{1/2}$. Hence, using rounded figures, one obtains

$$\epsilon_{12}/k = 1.276(T_{c,1}T_{c,2})^{0.453} \tag{10}$$

and

$$\sigma_{12} = 0.2947((V_{c,1}^{0.4} + V_{c,2}^{0.4}) \tag{11}$$

Combination of Equations 10 and 11 with Equation 5 yields (at 1-atm pressure)

$$D_{12} = \Gamma T^{3/2}/\Omega_{12}^{(1,1)*}(T_{12}^*) \tag{12}$$

where $\Gamma = 0.021397\ \mu^{-1/2}/(V_{c,1}^{0.4} + V_{c,2}^{0.4})$ is a constant characteristic of the system under consideration. As already pointed out (*1*), the advantage of Equation 12 over the original Chen-Othmmer relation (*2, 3*) lies in the fact that its temperature dependence is not always the same but may vary with the nature of the binary system.

Table III. Data Used in Calculations for Table I

Substance	σ, Å	(ϵ/k), K	I, eV	V_c, cm³ mol⁻¹ [a]	T_c, K[a]
SF_6	5.51	201	19.3	199.0	318.7
n-Hexane	5.92	517	10.43	369.9	507.4
n-Heptane	6.25	573	10.35	431.9	540.2
n-Octane	6.54	611	10.24	492.4	568.8
2,2,4-Trimethylpentane	6.52	602	10.0	468.2	543.9

[a] All critical data were taken from ref. *17*.

In both approaches (a) and (b), collision integrals were extracted from the tables of Hirschfelder et al. (*11*), which, for the relevant temperature range, are virtually identical with the more accurate tables of Monchik and Mason (*21*). Essentially the same features as with cyclic hydrocarbons-SF_6 were observed: whereas diffusion coefficients calculated via method (b) agreed quite well with experiment (cf. Table I), method (a) yielded substantially lower values, with deviations being as large as 16%. This corroborates the contention (*3*) that of all prediction methods utilizing semiempirical parameters, Chen and Othmer's method with its present modification is the most versatile approach, provided V_c and T_c are known. It is worthwhile noting that in both cases, the relative sequential order is reproduced satisfactorily, viz., $D_{C_6} > D_{C_7} > D_{C_8} \approx D_{\text{isooctane}}$.

Regularities in homologous series. According to the simple model of Humphreys and Mills (*13*), the following relation between D_{12} and μ should hold:

$$D_{12} \sim f(T)M_1^{1/2}\,\mu^{-1/2}/\left[\lambda^2 + 2\,\lambda\left(\frac{\mu}{M_1 - \mu}\right)^{1/3} + \left(\frac{\mu}{M_1 - \mu}\right)^{2/3}\right] \tag{13}$$

Here, λ is a constant for a particular homologous series which may be treated advantageously as a disposable fitting parameter (*28, 29*). Thus, under the provision that $f(T)$ does not vary within the series, the ratio of the diffusion coefficient of the *n*th member of the homologous series, in mixtures with a common gas G, to the diffusion coefficient of the first member of the series is solely a function of the reduced mass and should not depend on temperature (henceforward, for the sake of clarity, we denote the molar mass of the common gas by M_G instead of M_1, and that of the homolog by M_n, with $n = 1,2,3, \ldots$):

$$\frac{[D_{12}]_{n,G}}{[D_{12}]_{1,G}} = \left(\frac{\mu_1}{\mu_n}\right)^{1/2} \frac{\lambda^2 + 2\,\lambda q_1 + q_1^2}{\lambda^2 + 2\,\lambda q_n + q_n^2} \tag{14}$$

$$q_n = \left(\frac{M_G}{\mu_n} - 1\right)^{-1/3} \tag{15}$$

$$\mu_n = M_GM_n/(M_G + M_n) \tag{16}$$

A comparison of our data on the *n*-alkane series (cf. also ref. *34*) with the predictions of Equation 14 is displayed in Figure 1, where we have plotted experimental ratios $[D_{12}]_{n,G}/[D_{12}]_{1,G}$ against μ_n/M_G for a temperature of 298.15K and 1 atm. In addition to the pseudosphericals methane, carbon tetrafluoride, and sulfur hexafluoride, we included argon as a representative of an ideally spherical nonpolar common gaseous compound. The raw data have been smoothed with the aid of Equation 4 and the parameters of Table II.

Measurements for the lower alkanes are due to Mueller and Cahill (*22*) (CH_4-CH_4 and CH_4-CF_4), Weissman (*32*) (CH_4-C_2H_6 and CH_4-C_3H_8), and Gotoh et al. (*8*) [CH_4-*n*-C_4H_{10} and CH_4-SF_6; for the latter system, Kestin and Yata (*15*) report a slightly lower value]. The data for the argon systems up to *n*-C_4H_{10} were taken from ref. *29*. Diffusion coefficients for *n*-hexane, *n*-heptane, and *n*-octane in argon were estimated by using experimental results (*4, 5*) at 15° and 30°C, respectively.

By minimizing deviations from the empirical ratios $[D_{12}]_{n,G}/[D_{12}]_{1,G}$ the following figures were obtained at 298.15K (the subscript indicates the common gas): $\lambda_{CH_4} = 0.65$, $\lambda_{Ar} = 0.62$, $\lambda_{CF_4} = 1.07$, and $\lambda_{SF_6} = 0.47$. The rather narrow temperature range of our measurements does not allow any definite conclusions to be drawn as to a possible weak temperature dependence (*28, 29*) of λ. It is evident, that within a tolerance band of a few percent, *all* of the experimental data are reproduced. This substantiates the assertion (*28, 29*) that the hard sphere model correctly reflects the

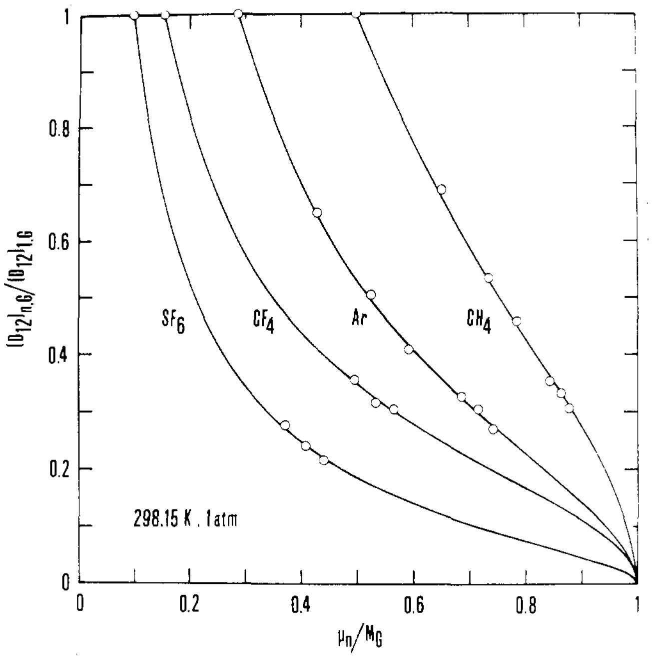

Figure 1. Ratio of binary gaseous diffusion coefficients, $[D_{12}]_{n,G}/[D_{12}]_{1,G}$, of *n*-alkanes in common gas (G = CH_4, Ar, CF_4, and SF_6, respectively), as function of dimensionless quantity μ_n/M_G at 298.15K and 1 atm (101.325 kPa)

Circles represent experimental ratios; solid curves signify predictions of Humphreys-Mills (*13*) model in form of Equation 14. Deviations of calculated from experimental values do not exceed in any case ±3%

observed regularities in the homologous series, although the quantitative success is somewhat surprising. Thus, Equation 14 may be profitably used as a powerful tool to predict binary diffusion coefficients in a homologous series with a common gas, provided that experimental data on two sufficiently spaced homologs are available.

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