New Generalized Equation for Gas Diffusion Coefficient

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THE IMPORTANCE of diffusion in chemical processes has been well known. In view of the scarcity of reliable experimental data for the diffusion coefficient, its value was often obtained from the correlation equations. During the past years, there were many investigations. The theoretical approach to this study was due to Fick (*9*), Stefan (*27*), Maxwell (*19*), Sutherland (*28*), Meyer (*20*), Jeans (*13*), Enskog (*7*), and Chapman and Cowling (*4*). The empirical or modified correlation equations have been developed by Arnold (*1*), Gilliland (*10*), Hirschfelder, Bird, and Spotz (*11*), Slattery (*24, 25*), and others (*8, 33*). The most reliable one is the Chapman and Cowlings' equation, which has been extensively investigated by Hirschfelder, Bird, and Spotz (*12*). Although the values calculated by the Chapman and Cowlings equation agree well with the experimental measurements, its use is somewhat cumbersome because it involves a long table for the collision integral function. For rapid calculations and for use in analytical solutions of the diffusion problem, a simpler and more generalized equation is still needed. The present investigation aimed to review briefly previous recent work, then to propose a new, simple, convenient, and more generalized equation for the gas diffusion coefficient of a binary system at low pressure, and finally to compare the result with those of previous investigators.

BRIEF REVIEW OF PREVIOUS CORRELATIONS

Arnold Method (*1*). Arnold, following Sutherland's general approach to use an additional term S_{12} to account for deviations from hard spheres, developed an empirical equation in which the numerical constant was evaluated from available experimental data. He proposed the following:

$$D_{12} = \frac{0.00837\, T^{3/2}\left(\frac{1}{M_1} + \frac{1}{M_2}\right)^{1/2}}{P(V_{b_1}^{1/3} + V_{b_2}^{1/3})^2\left(1 + \frac{S_{12}}{T}\right)} \tag{1}$$

where S_{12}, Sutherland's constant, is calculated from the following expression:

$$S_{12} = 1.47F(T_{b_1}T_{b_2})^{1/2} \tag{2}$$

in which

$$F = \left[\frac{2(V_{b_1}^{1/3}V_{b_2}^{1/3})^{1/2}}{V_{b_1}^{1/3} + V_{b_2}^{1/3}}\right]^3 \tag{3}$$

V_b is the molal volume at normal boiling point which can be estimated by the Le Bas method (*17*).

Gilliland Method (*10*). Gilliland adopted the hard sphere model of the classical kinetic theory to develop an empirical equation by assuming the collision diameters proportional to the cube roots of the molar volumes of the components at the normal boiling point as in the earlier method of Arnold. The numerical constant in the equation was also obtained from the available experimental data. His final equation is

$$D_{12} = \frac{0.0043\, T^{3/2}\left(\frac{1}{M_1} + \frac{1}{M_2}\right)^{1/2}}{P(V_{b_1}^{1/3} + V_{b_2}^{1/3})^2} \tag{4}$$

Hirschfelder, Bird, and Spotz Method (*11*). Hirschfelder, Bird, and Spotz, following the more rigorous kinetic theory as developed by Chapman and Cowling, have calculated the collision integral for the potential energy of attraction between two nonpolar gases in which the energy of attraction varies with the inverse sixth power of the distance between centers of adjacent molecules and the repulsive energy with the inverse twelfth power. The original equation as proposed by Chapman and Cowling to estimate the diffusion coefficient for binary pairs of spherical nonpolar molecules at low pressure is

$$D_{12} = \frac{0.001858\, T^{3/2}\left(\frac{1}{M_1} + \frac{1}{M_2}\right)^{1/2}}{P\,\sigma^2_{12}\,\Omega(1,1)^*} \tag{5}$$

The collision integral $\Omega(1,1)^*$ has been calculated by Hirschfelder, Bird, and Spotz as a function of $(kT/\epsilon)_{12}$ and listed in a long table. The values of ϵ/k and σ are obtained from viscosity data or estimated by the following expressions:

$$\frac{\epsilon}{k} = 0.77\, T_c \tag{6}$$

$$\sigma = 0.833\, V_c^{1/3} \tag{7}$$

$$\left(\frac{\epsilon}{k}\right)_{12} = \left[\left(\frac{\epsilon}{k}\right)_1 \left(\frac{\epsilon}{k}\right)_2\right]^{1/2} \tag{8}$$

$$\sigma_{12} = \frac{\sigma_1 + \sigma_2}{2} \tag{9}$$

Slattery and Bird Method (*24, 25*). Slattery and Bird recently developed an empirical equation involving the reduced temperature, critical temperature, and critical pressure as

$$D_{12} = \frac{aT_r^b(P_{c_1}P_{c_2})^{1/3}(T_{c_1}T_{c_2})^{5/12}\left(\frac{1}{M_1} + \frac{1}{M_2}\right)^{1/2}}{P} \tag{10}$$

where

$$T_r = \frac{T}{(T_{c_1}T_{c_2})^{1/2}}$$

$a = 2.74 \times 10^{-4}$ for interdiffusion of nonpolar gases and for self diffusion
$= 3.63 \times 10^{-4}$ for interdiffusion of water and a nonpolar gas
$b = 1.823$ for interdiffusion of nonpolar gases and for self-diffusion
$= 2.334$ for interdiffusion of water and a nonpolar gas

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DEVELOPMENT OF NEW GENERALIZED EQUATION

Relationship between ϵ/k and T_c, σ and V_c. Hirschfelder, Bird, and Spotz proposed to calculate the force constants by Equations 6 through 9 when the force constants are not available in the table from viscosity data. However, the relationship between ϵ/k and T_c and that between σ and V_c were tested, with selected gases (*12, 21*) as listed in Table I, by the method of least squares (*30*) as

$$\epsilon/k = 1.276\ T_c^{0.9061} \qquad (11)$$

$$\sigma = 0.5894\ V_c^{0.4006} \qquad (12)$$

These relationships as well as those proposed by Hirschfelder, Bird, and Spotz were plotted in Figures 1 and 2. It is obvious that Equations 11 and 12 correlate data better than Equations 6 and 7.

Elimination of Use of Collision Integral Table. Hirschfelder, Bird, and Spotz have calculated the value of $\Omega(1,1)^*$ as function $(kT/\epsilon)_{12}$ in a long table to be used in Equation 5 as developed by Chapman and Cowling. This lengthy table was reduced to an equation by Chen (*5*) as

$$\Omega(1,1)^* = \frac{1.075}{\left(\dfrac{kT}{\epsilon}\right)_{12}^{0.1615}} + \frac{2}{\left(10\dfrac{kT}{\epsilon}\right)_{12}^{0.74\log_{10}\left(10\frac{kT}{\epsilon}\right)_{12}}} \qquad (13)$$

The accuracy of this equation from the original table was reported (*5*) with an average deviation of −0.078% from −1.92% to +0.53% for 82 point values.

Now substitution of ϵ/k from Equation 11 into Equation 8 and then of the resulting $(\epsilon/k)_{12}$ into Equation 13 yields

$$\Omega(1,1)^* = \frac{1.075}{\left[\dfrac{T}{1.276(T_{c_1}T_{c_2})^{0.453}}\right]^{0.1615}} + \frac{2}{\left[\dfrac{10T}{1.276(T_{c_1}T_{c_2})^{0.453}}\right]^{0.74\log_{10}\left[\frac{10T}{1.276(T_{c_1}T_{c_2})^{0.453}}\right]}} \qquad (14)$$

or

$$\Omega(1,1)^* = f\left[\frac{T}{(T_{c_1}T_{c_2})^{0.453}}\right] \qquad (15)$$

By combining the σ_{12} obtained from Equation 12 into Equation 9 with Equations 15 and 5 we get:

$$D_{12} = \frac{0.001858\ T^{3/2}\left(\dfrac{1}{M_1}+\dfrac{1}{M_2}\right)^{1/2}}{P\,0.2947[(V_{c_1}^{0.4006}+V_{c_2}^{0.4006})]^2\ f\left[\dfrac{T}{(T_{c_1}T_{c_2})^{0.453}}\right]} \qquad (16)$$

For approximation if we assume:

$$f\left[\frac{T}{(T_{c_1}T_{c_2})^{0.453}}\right] = A\left[\frac{T}{(T_{c_1}T_{c_2})^{0.453}}\right]^B \qquad (17)$$

and use $V_c^{0.4}$ instead of $V_c^{0.4006}$, Equation 16 becomes

$$\frac{T^{3/2}\left(\dfrac{1}{M_1}+\dfrac{1}{M_2}\right)^{1/2}}{P(V_{c_1}^{0.4}+V_{c_2}^{0.4})^2 D_{12}} = A\left[\frac{T}{(T_{c_1}T_{c_2})^{0.453}}\right]^B \qquad (18)$$

Table I. Selected Gases for Figures 1 and 2

Acetylene
Air
Ammonia
Argon
Benzene
Carbon dioxide
Carbon disulfide
Carbon monoxide
Carbon tetrachloride
Chloroform
Chlorine
Cyclohexane
Ethane
Ethylene
Ethanol
Ethyl ether
Freon-12
Helium
n-Hexane
Hydrogen
Hydrogen chloride
Krypton
Methane
Methanol
Neon
Nitrogen
Nitrous oxide
Nitric oxide
Oxygen
n-Pentane
Sulfur dioxide
Water
Xenon

T_c and V_c from (*21*); ϵ/k and σ from (*12*).

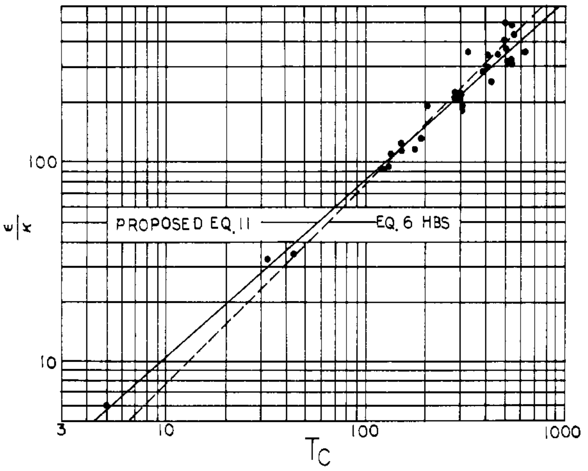

Figure 1. ϵ/k vs. T_c

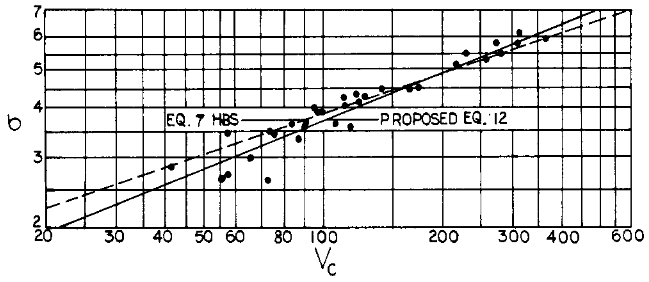

Figure 2. σ vs. V_c

Evaluation of A' and B'. The next step is to evaluate A' and B' from the available experimental data. For convenience of computation, two groups of variables

$$\frac{\dfrac{T}{100}}{\left(\dfrac{T_{c_1}T_{c_2}}{10{,}000}\right)^{0.453}} \quad \text{and} \quad \frac{\left(\dfrac{T}{100}\right)^{3/2}\left(\dfrac{1}{M_1}+\dfrac{1}{M_2}\right)^{1/2}}{PD_{12}\left[\left(\dfrac{V_{c_1}}{100}\right)^{0.4}+\left(\dfrac{V_{c_2}}{100}\right)^{0.4}\right]^2}$$

for the first 66 (No. 1 to 66) different binary gaseous systems as listed in Table II were calculated. By plotting these two groups of variables on log log coordinate paper, the values of modified A' and B' can be determined by the method of least squares. Figure 3 is such a plot. The values of modified A' and B' thus determined are 0.3666 and −0.3102, respectively. The correlation coefficient of this straight line calculated by means of statistics (*30*) is −0.9452. This means

Table II. Selected Binary System for Correlation

Plot (Nos. 1 to 66) and for Comparison (Nos. 6 to 71)

	System	I T, ° K.	II PD_{12}, Exptl.	III Lit., Ref.	IV PD_{12}, Calcd.	V %, Deviation
1.	Carbon dioxide–ethyl acetate	319	0.0666	(*22*)		
2.	Air–ethyl acetate	273	0.0709	(*1*)		
3.	Air–ethyl butyrate	273	0.0574	(*1*)		
4.	Air–propyl acetate	315	0.0920	(*1*)		
5.	Air–toluene	298	0.0844	(*1*)		
6.	Air-ammonia	273	0.1980	(*1*)	0.1960	−1.01
7.	Air-benzene	298	0.0962	(*16*)	0.0890	−7.49
8.	Air–carbon dioxide	273	0.1360	(*22*)	0.1430	+5.15
9.	Air–carbon disulfide	273	0.0883	(*1*)	0.0925	+5.15
10.	Air-chlorine	273	0.1240	(*22*)	0.1128	−9.04
11.	Air–ethanol	298	0.1320	(*22*)	0.1210	−8.34
12.	Air–ethyl ether	293	0.0896	(*29*)	0.0859	−4.13
13.	Air–methanol	298	0.1620	(*22*)	0.1530	−5.55
14.	Air–*n*-octane	298	0.0602	(*18*)	0.0618	+2.66
15.	Air-oxygen	273	0.1750	(*22*)	0.1860	+6.30
16.	Air–sulfur dioxide	273	0.1220	(*22*)	0.1150	−5.74
17.	Air-water	298	0.2600	(*16*)	0.2340	−10.00
18.	Ammonia-ethylene	293	0.1770	(*28*)	0.1670	−5.65
19.	Argon-neon	293	0.3290	(*28*)	0.3420	+3.95
20.	Carbon dioxide-benzene	318	0.0715	(*22*)	0.0736	+2.94
21.	Carbon dioxide–carbon disulfide	318	0.0715	(*22*)	0.0895	+25.20
22.	Carbon dioxide–ethanol	273	0.0693	(*14*)	0.0733	+11.50
23.	Carbon dioxide–ethyl ether	273	0.0541	(*28*)	0.0558	+3.14
24.	Carbon dioxide–hydrogen	273	0.5500	(*4*)	0.5750	+4.54
25.	Carbon dioxide–methane	273	0.1530	(*4*)	0.1540	+0.65
26.	Carbon dioxide–methanol	298.6	0.1050	(*22*)	0.1170	+11.40
27.	Carbon dioxide–nitrogen	298	0.1580	(*22*)	0.1650	+4.43
28.	Carbon dioxide–nitrous oxide	298	0.1170	(*32*)	0.1240	+5.98
29.	Carbon dioxide–water	298	0.1640	(*29*)	0.1820	+11.00
30.	Carbon monoxide–ethylene	273	0.1510	(*29*)	0.1380	−8.61
31.	Carbon monoxide–hydrogen	273	0.6510	(*22*)	0.6690	+2.76
32.	Carbon monoxide–nitrogen	288	0.1920	(*22*)	0.1970	+2.60
33.	Carbon monoxide–oxygen	273	0.1850	(*29*)	0.1800	−2.70
34.	Freon 12–benzene	298	0.0385	(*16*)	0.0365	−5.20
35.	Freon 12–ethanol	298	0.0475	(*16*)	0.0529	+11.40
36.	Freon 12–water	298	0.1050	(*16*)	0.1068	+1.71
37.	Helium-argon	273	0.6410	(*4*)	0.7000	+9.20
38.	Helium-ethanol	298	0.4940	(*16*)	0.4740	−4.05
39.	Helium-benzene	298	0.3840	(*16*)	0.3740	−2.60
40.	Helium-hydrogen	293	1.6400	(*29*)	1.7200	+4.88
41.	Helium-neon	293	1.2300	(*29*)	1.2400	+0.81
42.	Helium-water	298	0.9080	(*16*)	0.7900	−13.00
43.	Hydrogen-ammonia	293	0.8490	(*29*)	0.7340	−13.55
44.	Hydrogen-argon	293	0.7700	(*31*)	0.8050	+4.54
45.	Hydrogen-benzene	273	0.3170	(*29*)	0.3290	+3.78
46.	Hydrogen–carbon tetrachloride	296	0.3450	(*29*)	0.3670	+6.38
47.	Hydrogen-ethane	273	0.4390	(*29*)	0.4840	+10.20
48.	Hydrogen-ethanol	340	0.5780	(*29*)	0.6190	+7.10
49.	Hydrogen-ethylene	298	0.6020	(*22*)	0.6090	+1.26
50.	Hydrogen–ethyl ether	293	0.3540	(*29*)	0.3680	+3.96
51.	Hydrogen–hydrogen chloride	294	0.7950	(*29*)	0.6870	−13.50
52.	Hydrogen-methane	273	0.6250	(*4*)	0.6260	+0.16
53.	Hydrogen-nitrogen	293	0.7600	(*22*)	0.7740	+1.84
54.	Hydrogen–nitrous oxide	273	0.5350	(*4*)	0.5680	+6.16
55.	Hydrogen-oxygen	273	0.6970	(*4*)	0.7140	+2.44
56.	Hydrogen–sulfur dioxide	273	0.4830	(*14*)	0.4880	+1.03
57.	Hydrogen-water	293	0.8500	(*29*)	0.7550	−11.10
58.	Nitrogen-ammonia	293	0.2410	(*29*)	0.2200	−8.70
59.	Nitrogen-ethylene	298	0.1630	(*22*)	0.1630	0
60.	Nitrogen-hydrogen	288	0.7430	(*3*)	0.7490	+0.81
61.	Nitrogen–nitric oxide	293	0.2320	(*29*)	0.2280	−1.72
62.	Nitrogen-oxygen	273	0.1810	(*4*)	0.1820	+0.55
63.	Oxygen-ammonia	293	0.2530	(*29*)	0.2240	−11.10
64.	Oxygen-benzene	296	0.0939	(*29*)	0.0862	−8.20
65.	Oxygen–carbon tetrachloride	298	0.0731	(*29*)	0.0780	+6.70
66.	Oxygen-ethylene	293	0.1820	(*29*)	0.1600	−12.10
67.	Carbon dioxide–propane	298	0.0863	(*32*)	0.0885	+2.55
68.	Ethane-propane	293	0.0850	(*29*)	0.0832	−2.12
69.	Ethane-methane	293	0.1630	(*29*)	0.1570	−3.68
70.	Hydrogen-propane	300	0.4500	(*29*)	0.4680	+4.00
71.	Nitrous oxide–propane	298	0.0860	(*32*)	0.0880	+2.32
					Av. abs. dev.	5.64

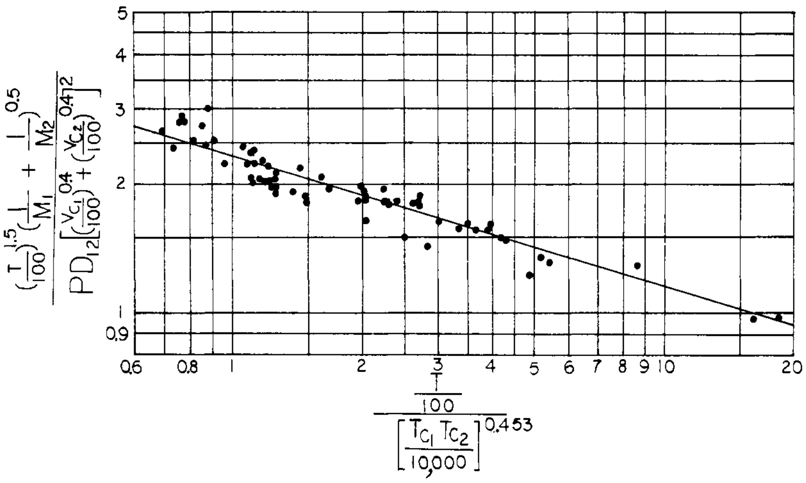

Figure 3. Proposed correlation plot

that the data are highly correlated. Consequently, Equation 18 becomes

$$\frac{\left(\frac{T}{100}\right)^{3/2}\left(\frac{1}{M_1}+\frac{1}{M_2}\right)^{1/2}}{PD_{12}\left[\left(\frac{V_{c1}}{100}\right)^{0.4}+\left(\frac{V_{c2}}{100}\right)^{0.4}\right]^2} = 0.3666\left[\frac{\left(\frac{T}{100}\right)}{\left(\frac{T_{c1}T_{c2}}{10,000}\right)^{0.453}}\right]^{-0.3102} \qquad (19)$$

Simplification of Equation 19 gives

$$D_{12} = \frac{0.43\left(\frac{T}{100}\right)^{1.81}\left(\frac{1}{M_1}+\frac{1}{M_2}\right)^{0.5}}{P\left(\frac{T_{c1}T_{c2}}{10,000}\right)^{0.1405}\left[\left(\frac{V_{c1}}{100}\right)^{0.4}+\left(\frac{V_{c2}}{100}\right)^{0.4}\right]^2} \qquad (20)$$

COMPARISON OF RESULTS

The last 66 (Nos. 6 to 71) binary systems in Table II were selected for the determination of the diffusion coefficient by the proposed Equation 20. The calculated values and per cent deviation were shown in columns IV and V (Table II), respectively. These same 66 binary systems were calculated by the Arnold method, Gilliland method, and Hirschfelder, Bird, and Spotz, method (called HBS method) with estimated force constants, with force constants obtained from viscosity data, and by the Slattery method. These calculations were tabulated in detail in (*6*) and the comparison is shown in Table III.

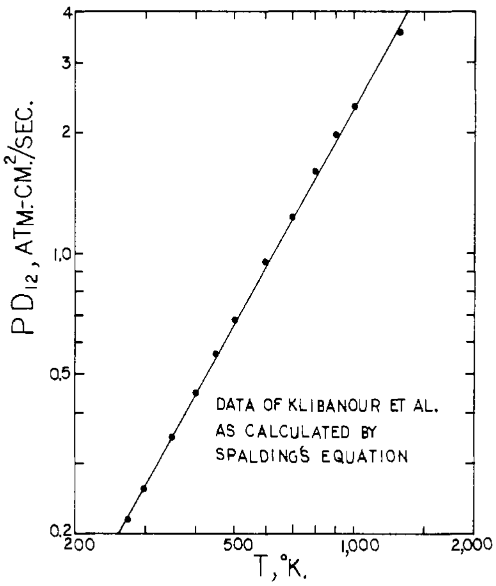

Figure 4. Diffusion coefficient of water-air at different temperatures

EFFECT OF TEMPERATURE ON DIFFUSION COEFFICIENT

Theory and experiment indicate that the diffusion coefficient varies inversely with pressure and directly with temperature as a power function. This temperature coefficient as defined by Benedict (*2*) is not the same for all the binary systems. The average value has been described as 1.50 by the Gilliland method, 1.823 for nonpolar gases, 2.334 for water and a nonpolar gas by the Slattery and Bird method, 1.81 by the proposed method, a complicated function by the Arnold method and the Hirschfelder, Bird, and Spotz method. The verification of the proposed value 1.81 follows.

The experimental data of Klibanour, Pomerantsev, and Frank-Kamenetsky (*15*) on water vapor in air have been reduced to an equation by Spalding (*26*) and described by Sherwood and Pigford (*23*). Hence the diffusion coefficient of water vapor in air at different temperatures up to 2000° F. can be calculated. By plotting these diffusion coefficients at the corresponding temperature in Figure 4, it is seen that the slope of the straight line is about 1.82, which agrees very well with the proposed value of 1.81 and differs widely from the 1.50 value of Gilliland and 2.334 value of Slattery and Bird.

Another set of experimental data (*12*) for the diffusion coefficient of binary systems at various temperatures was tested by the previous five correlation methods and the proposed method. It was shown in Table XV (*6*) that the average absolute deviation is 2.00% by the proposed method, 3.83% by the Slattery and Bird method, 5.17% by

Table III. Comparison of Results by Different Methods

Method	Arnold	Gilliland	HBS, Estimated	HBS, Viscosity	Slattery	Present Author
Equations	1 to 3	4	5 to 9	5	10	20
Av. abs. dev., %	16.01	16.80	11.23	6.12	9.90	5.64
Av. dev., %	9.85	−10.80	−10.19	−2.56	+1.11	+0.22
Std. dev. (S.D.), %	±22.49	±16.97	±8.50	±7.46	±20.35	±7.21
% sample within 1 S.D.	78.79	60.61	41.00	66.60	92.50	71.21
Lower max. dev., %	−37.40	−60.20	−30.10	−17.90	−23.00	−13.55
Upper max. dev., %	+90.50	+27.30	+9.10	+13.10	+137.00	+25.20
Abs. max. dev., %	90.50	60.20	30.10	17.90	137.00	25.20

the HBS method (with force constants), 9.84% by the HBS method (with estimated force constants), 19.15% by the Gilliland method, 12.91% by the Arnold method. The comparison of these results for the effect of temperature on diffusion coefficient was tabulated in Table XV (*6*).

This temperature coefficient was recorded as either 1.75 or 2.00 by Benedict (*2*). For 65 binary systems in the table of this reference, the average value is 1.89, which agrees closely with the proposed value 1.81 and differs appreciably from 1.50 of Gilliland. Although it agrees more closely with the value 1.823 of Slattery and Bird, other effects, such as molecular weight and collision diameter, on the diffusion coefficient have shown that the proposed method is better than the Slattery and Bird method.

CONCLUSIONS

From Table III, it is readily seen that the proposed Equation 20 gives the lowest values for average absolute deviation, average deviation, standard deviation, and lower maximum deviation. As shown in the same table, the results of Arnold, Gilliland, and HBS with estimated force constants are evidently not as accurate as those obtained by the proposed Equation 20. The Slattery method shows the highest percentage of sample within one standard deviation; however, the standard deviation in the Slattery method is about three times as large as that by the proposed Equation 20 and the upper maximum deviation is about five times as large as that by Equation 20, the lower maximum deviation is about 1.7 times as large as that by Equation 20. From these statistical analyses, it is also seen that the author's method is better than the Slattery method. The proposed Equation 20 is still somewhat better than the HBS method, using force constants from viscosity data (Equation 5). The other disadvantage in using Equation 5 is that the force constants from viscosity data are not adequate for the estimation of the diffusion coefficient of a binary mixture, whereas the proposed Equation 20 requires critical data which are available.

Because of the simplicity, accuracy, and convenience in the use of Equation 20, it is proposed to use it for the estimation of the gas diffusion coefficient of a binary mixture at low pressure. This proposed equation is valid within the range of 0.7 to about 20 of the following:

$$\frac{\dfrac{T}{100}}{\left(\dfrac{T_{c_1} T_{c_2}}{10{,}000}\right)^{0.453}}$$

The proposed temperature coefficient 1.81 was verified as reasonable and more reliable than those obtained by previous investigators.

NOMENCLATURE

a = numerical constant in Equation 10
A = numerical constant in Equation 17
A' = modified numerical constant
b = numerical constant in Equation 10
B = numerical constant in Equation 17
B' = modified numerical constant
D = diffusion coefficient, sq. cm. per sec.
ϵ = maximum energy of molecular attraction, ergs
F = a factor in Equation 2
k = Boltzmann's constant, ergs per ° K.
M = molecular weight
P = pressure, atm.
S.D. = standard deviation
S_{12} = Sutherland's constant, ° K.
T = temperature, ° K.
T_r = reduced temperature, ° K. = $T/(T_{c_1} T_{c_2})^{1/2}$
V = molal volume, cc. per gram mole
σ = collision diameter, A.
$\Omega(1, 1)^*$ = collision integral function for molecular diffusion

Subscripts

b = at the normal boiling point
c = at the critical point
1 or 2 = component 1 or 2
12 = for a binary system

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