Prediction of Diffusion Coefficients for Nonelectrolytes in Dilute Aqueous Solutions

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Diffusivities of dissolved substances in water have been correlated by Wilke and Chang (1955), Scheibel (1954), and Othmer and Thakar (1953). These correlations have proven to be fairly reliable yielding similar, but by no means identical, results.

Othmer and Thakar:

$$D_{12} = \frac{14.0(10^{-5})}{\mu_2^{1.1} V_1^{0.6}} \tag{1}$$

Wilke and Chang:

$$D_{12} = \frac{7.4(10^{-8})(2.6\,M_2)^{0.5}\,T}{\mu_2 V_1^{0.6}} \tag{2}$$

Scheibel:

For $V_1 > V_2$:

$$D_{12} = \frac{8.2(10^{-8})[1 + (3\,V_2/V_1)^{2/3}]\,T}{\mu_2 V_1^{1/3}} \tag{3a}$$

If $V_1 < V_2$:

$$D_{12} = \frac{25.2(10^{-8})\,T}{\mu_2 V_1^{1/3}} \tag{3b}$$

All three correlations were developed from data largely obtained prior to 1950 and included that from the *International Critical Tables* (1929) and the work of Arnold (1930). Reid and Sherwood (1966) analyzed these correlations for water as solvent and found that all three gave identical average absolute errors of 11%. It is noted, though, that the analysis was based on diffusivities of some 37 substances of which 15 were obtained from the *International Critical Tables.* It appeared of considerable interest, therefore, to determine the extent to which the correlations were still applicable in the light of the large number of diffusivities reported since 1950. For the purpose of mathematically testing the three correlations, only data appearing in the literature since 1950 were compiled. In some instances even relatively recent data were judged inaccurate when compared with those of other workers and were excluded. The mathematical analysis initially took the form of determining average errors between measured diffusivities and those estimated from the three correlations.

Subsequently it was possible to propose new constants for the Othmer-Thakar equation, and a new value for the association parameter for the Wilke-Chang equation when applied to water as solvent. The revised Othmer-Thakar equation based on diffusivities of some 87 different dissolved substances in dilute aqueous solutions is

$$D_{12} = \frac{13.26(10^{-5})}{\mu_2^{1.4} V_1^{0.589}} \tag{4}$$

The revised association parameter for water of 2.26 instead of 2.6 is likewise recommended for use in the Wilke-Chang equation.

Also reported in this work are the diffusivities of vinyl chloride monomer in water which were measured at atmospheric pressure and temperatures of 25°, 50°, and 75°C using the steady state capillary cell method previously described by Malik and Hayduk (1968) and Hayduk and Cheng (1971).

New developments in liquid diffusion have been recently reviewed by Ghai et al. (1973) and Ertl et al., (1974).

EXPERIMENT

Distilled water and vinyl chloride having a specified minimum purity of 99.9% purchased from Matheson of Canada, were used in these experiments. The calculation of diffusivity depended on a knowledge of equilibrium solubilities of vinyl chloride in water which have been measured in this laboratory and reported elsewhere (Hayduk and Laudie, 1974).

Integral binary diffusivities were calculated from the steady state rate of shrinkage of the gas confined within the capillaries of the diffusion cell. The equilibrium concentration at atmospheric pressure was sufficiently low, less than 0.002 mole fraction, so that the integral diffusivity was considered to approximate the diffusivity at infinite dilution. The appropriate equation for calculating the diffusivity for solutions of constant mass concentration was

$$D = \frac{n_A L}{\rho \ln\left(\dfrac{1 + \omega_{A0}}{1 + \omega_{AL}}\right)} \tag{5}$$

The mass flux, length of diffusion path, and solution density (taken as solvent density) were measured or were readily calculated. The vinyl chloride concentration at the gas-liquid interface was calculated with due consideration for the barometric pressure and solvent partial pressure. The vinyl chloride concentration within the diffusion cell was always calculated to be less than 1% saturated.

The average of at least three results for each of the vinyl chloride diffusivities in water is compared with predicted values for 25°C in Table 1. The reproducibility for these measurements was usually greater than 3%, and the accuracy was estimated at better than 4%.

DISCUSSION

Selected diffusivities in water at 25°C and sources are listed in Table 2 along with molar volumes as estimated by the LeBas incremental method as described in Perry (1964), and where possible, as determined from liquid densities. In some instances diffusivities at 25°C were obtained by extrapolation of data at other temperatures. In his review Himmelblau (1964) had suggested that

TABLE 1. DIFFUSIVITY OF VINYL CHLORIDE IN WATER

		D, cm²/s (× 10⁵)				
Temp., °C	$(10^4)x_2$*	mean	Othmer & Thakar (1953)	Scheibel (1954)	Wilke & Chang (1955)	Equation (4)
25	7.98	1.34	1.30	1.31	1.39	1.30
50	4.10	2.42				
75	2.25	3.67				

* Hayduk and Laudie (1974).

TABLE 2. DIFFUSIVITIES IN WATER AT 25°C AND SOURCES

No.	Substance	V_2		$D \times 10^5$	Sources for diffusivities
		LeBas	Actual		
1.	H_2	14.3	28.5^a	4.40	Ferrell and Himmelblau, 1967
2.	He	—	31.9^a	6.28	Ferrell and Himmelblau, 1967
3.	NO	23.6	23.6^b	2.34	Wise and Houghton, 1968
4.	N_2O	36.4	36.0^a	2.10	Thomas and Adams, 1965
5.	N_2	31.2	34.7^a	1.99	Ferrell and Himmelblau, 1967
6.	NH_3	25.8	24.5^a	2.28	Himmelblau, 1964
7.	O_2	25.6	27.9^a	2.29	Ferrell and Himmelblau, 1967
8.	CO	30.7	34.5^a	2.30	Wise and Houghton, 1968
9.	CO_2	34.0	37.3^a	1.92	Ferrell and Himmelblau, 1967
10.	SO_2	44.8	43.8^a	1.59	Groothuis and Kramers, 1955
11.	H_2S	32.9	35.2^a	2.10	Himmelblau, 1964
12.	Cl_2	48.4	45.5^a	1.48	Vivian and Peaceman, 1956
13.	Ar	—	29.2^a	1.98	Baird and Davidson, 1962
14.	Kr	—	34.9^c	1.92	Wise and Houghton, 1968
15.	Ne	—	16.8^c	3.01	Houghton et al, 1962
16.	CH_4	29.6	37.7^b	1.67	Witherspoon and Bonoli, 1969
17.	CH_3Cl	47.5	50.6^a	1.49	Chiang and Toor, 1960
18.	C_2H_3Cl	62.3	64.5	1.34	This work
19.	C_2H_6	51.8	53.5^c	1.38	Witherspoon and Bonoli, 1969
20.	C_2H_4	44.4	49.4^b	1.55	Huq and Wood, 1968
21.	C_3H_8	74.0	74.5^b	1.16	Witherspoon and Bonoli, 1969
22.	C_3H_6	66.6	69.0^a	1.44	Vivian and King, 1964
23.	C_4H_{10}	96.2	96.6^d	0.97	Witherspoon and Saraf, 1965
24.	Pentane	118	118^d	0.97	Witherspoon and Bonoli, 1969
25.	C-pentane	99.5	97.9^e	1.04	Bonoli and Witherspoon, 1968
26.	C-hexane	118	117^b	0.90	Bonoli and Witherspoon, 1968
27.	M-c-pentane	112	120^e	0.93	Bonoli and Witherspoon, 1968
28.	Benzene	96.0	96.5^b	1.09	Ratcliff and Reid, 1961
29.	Toluene	118	118^d	0.95	Bonoli and Witherspoon, 1968
30.	E-benzene	140	140^d	0.90	Bonoli and Witherspoon, 1968
31.	Methanol	37.0	42.5^b	1.66	Gary-Bobo and Weber, 1969
32.	Ethanol	59.2	62.6^e	1.24	Hammond and Stokes, 1953
33.	Propanol	81.4	81.8^b	1.12	Gary-Bobo and Weber, 1969
34.	I-propanol	81.4	81.8^e	1.08	Gary-Bobo and Weber, 1969
35.	Butanol	104	101^e	0.98	Lyons and Sandquist, 1953
36.	I-butanol	104	101^e	0.93	Garner and Marchant, 1961
37.	N-butanol	104	102^e	0.97	Gary-Bobo and Weber, 1969
38.	Benzyl alcohol	126	121^e	0.93	Garner and Marchant, 1961
39.	E-glycol	66.6	63.5^f	1.16	Byers and King, 1966
40.	Tri-e-glycol	170	163^f	0.76	Merliss and Colver, 1969
41.	Pro-glycol	88.8	84.5^f	1.00	Garner and Marchant, 1961
42.	Glycerol	96.2	85.1^e	0.93	Garner and Marchant, 1961
43.	Ethane-hex-diol	200	197^g	0.64	Garner and Marchant, 1961
44.	Acetone	74.0	77.5^b	1.28	Anderson et al, 1958
45.	E-acetate	111	106^b	1.12	Chandrasekaran and King, 1972
46.	Furfural	95.7	97.5^e	1.12	Lewis, 1955
47.	Water (self)	18.9	18.8	2.45	Robinson and Stokes, 1955
48.	Urea	58.0	^{h}	1.38	Gosting and Akeley, 1952
49.	Formamide	43.8	46.6^e	1.67	Gary-Bobo and Weber, 1969
50.	Acetamide	66.0	67.1^g	1.32	Gary-Bobo and Weber, 1969
51.	Propionamide	88.2	87.4^e	1.20	Gary-Bobo and Weber, 1969
52.	Butyramide	110	109^g	1.07	Gary-Bobo and Weber, 1969
53.	I-butyramide	110	103^g	1.02	Gary-Bobo and Weber, 1969
54.	Diethylamine	112	109^b	1.11	Lewis, 1955
55.	Aniline	110	106^e	1.05	Lewis, 1955
56.	Formic acid	41.6	41.1^e	1.52	Bidstrup and Geankoplis, 1963
57.	Acetic acid	63.8	64.1^b	1.19	Lewis, 1955
58.	Propionic acid	86.0	85.3^e	1.01	Albery et al, 1967
59.	Butyric acid	108	108^e	0.92	Bidstrup and Geankoplis, 1963
60.	I-butyric acid	108	109^b	0.95	Albery et al., 1967
61.	Valeric acid	130	129^e	0.82	Bidstrup and Geankoplis, 1963
62.	Tri-m-acetic acid	130	127^g	0.82	Albery et al., 1967
63.	Chloracetic acid	81.7	79.6^e	1.04	Garland and Stockmayer, 1965
64.	Glycolic acid	71.2	^{h}	0.98	Albery et al., 1967
65.	Caproic acid	153	^{h}	0.78	Bidstrup and Geankoplis, 1963
66.	Succinic acid	120	^{h}	0.86	Albery et al., 1967
67.	Glutaric acid	142	^{h}	0.79	Albery et al., 1967

68.	Adipic acid	165	h	0.74	Albery et al., 1967
69.	Pimelic acid	187	h	0.71	Albery et al., 1967
70.	Glycine	78.0	h	1.06	Longsworth, 1952-1953
71.	Alanine	100	h	0.91	Longsworth, 1953
72.	Serine	108	h	0.88	Longsworth, 1953
73.	Aminobutyric acid	122	h	0.83	Longsworth, 1953
74.	Valine	145	h	0.77	Longsworth, 1953
75.	Leucine	167	h	0.73	Longsworth, 1953
76.	Proline	127	h	0.88	Longsworth, 1953
77.	Hydroxy-proline	135	h	0.83	Longsworth, 1953
78.	Histidine	168	h	0.73	Longsworth, 1953
79.	Phenylalanine	189	h	0.705	Longsworth, 1953
80.	Tryptophan	223	h	0.660	Longsworth, 1953
81.	Diglycine	138	h	0.791	Longsworth, 1953
82.	Triglycine	198	h	0.665	Longsworth, 1953
83.	Glycyl-leucine	227	h	0.623	Longsworth, 1953
84.	Leucyl-glycine	227	h	0.613	Longsworth, 1953
85.	Leucyl-glycyl-glycine	287	h	0.551	Longsworth, 1953
86.	Amino benzoic acid	144	h	0.840	Longsworth, 1953-54
87.	Dextrose	166	h	0.675	Gladden and Dole, 1953
88.	Sucrose	325	h	0.524	Henrion, 1964
89.	Raffinose	480	h	0.434	Longsworth, 1953

Sources for Molar volume or density data: *a.* Himmelblau, 1964; *b.* Reid and Sherwood, 1966; *c.* Bell, 1963; *d.* Rossini, 1953; *e.* Timmermans, 1950; *f.* Gallant, 1967; *g.* Weast, 1971; *h.* substance sublimes or decomposes.

true molar volumes should be more closely related to the diffusion process and hence yield better correlations than those based on LeBas molar volumes. To determine the validity of this consideration, the three previously mentioned correlations were tested using both molar volumes when this was possible. The LeBas method for oxygen in carboxylic acids was interpreted as suggested by Bidstrup and Geankoplis (1963) in which the contribution to the molar volume of the double bonded oxygen was considered as 12.0 cc/mole and of the oxygen in the hydroxyl group as 7.4 cc/mole.

The accuracy of the Wilke-Chang, Scheibel and Othmer-Thakar correlations was tested by calculating for each an average error (E_{avg}), average absolute error (E_{abs}), and a maximum error (E_{max}) based on diffusivities for all the substances listed in Table 2 excepting H_2 and He. These error estimates are listed in Table 3. Of the three, the Othmer-Thakar correlation gave the smallest error while the Wilke-Chang correlation gave diffusivities which tended to be too high.

An attempt was made to modify the Wilke-Chang correlation to conform to the recent data. Bidstrup and Geankoplis (1963) had formerly considered that the Wilke-Chang correlation predicted diffusivities that were 11% too high and suggested a revision of the constant from $7.4(10^{-8})$ to $6.6(10^{-8})$, based on work for the diffusivities of carboxylic and α-amino carboxylic acids. This work, based on a much larger number of substances would suggest an average reduction of 6.9%. It should be clarified, however, that the Wilke-Chang correlation was not tested in this work (nor that of Bidstrup and Geankoplis) for diffusivities in solvents other than water so that there is little justification for changing the constant in that equation. The association parameter is specific to water, however, so that predicted diffusivities in water can be suitably adjusted by reducing the value of this parameter from 2.6 to 2.26. The latter value for the association parameter yields low average errors as shown in Table 3. It is not possible to modify the Scheibel correlation for diffusivities in water by a similar adjustment to a constant.

TABLE 3. COMPARISON OF CORRELATIONS FOR DIFFUSIVITIES OF DILUTE SOLUTES IN WATER AT 25°C

	V_2 LeBas			V_2 Actual (where possible)		
	E_{avg}	E_{abs}	E_{max}	E_{avg}	E_{abs}	E_{max}
Wilke-Chang	7.2	8.8	33.4[a]	6.9	8.6	33.4[a]
Scheibel	2.5	6.7	29.0[b]	2.0	6.4	29.2[b]
Othmer-Thakar	0.5	5.9	25.1[a]	0.2	6.0	25.1[a]
Wilke-Chang ($X = 2.26$)	0.0	5.8	24.4[a]	-0.4	5.9	24.4[a]
This work [Equation (4)]	0.7	5.8	24.9[a]	0.3	5.9	24.9[a]

[a] For diffusivity of pentane.
[b] For self-diffusivity of water.

In an effort to determine whether an even better representation of the diffusivity data could be obtained, the constants in the Othmer-Thakar equation were reevaluated. To determine a suitable temperature coefficient of diffusivity, data were plotted as a function of the corresponding solvent viscosity as shown in Figure 1. For all the substances shown (and for many other substances not shown) of widely varying molecular size, log D was found to vary linearly with log μ with lines having an average slope determined as -1.14. For comparison in Figure 2, log D is also shown for the same substances as a function of log (T/μ). The resulting relation is required to be linear with a slope of 1 for application to both the Wilke-Chang and Scheibel correlations. By comparison of such plots it was concluded that the temperature coefficient was adequately represented by either a simple exponential viscosity function with no additional explicit temperature term or the T/μ function as in the Wilke-Chang and Scheibel correlations. The simpler of the two functions appeared preferable as a basis for correlation. Subsequently a linear regression of log D — log V_1 for diffusivities at 25°C yielded the revised constants as indicated in Equation (4). The accuracy of Equation (4) for predictive purposes was tested in the same manner as previously mentioned, the results of which are also shown

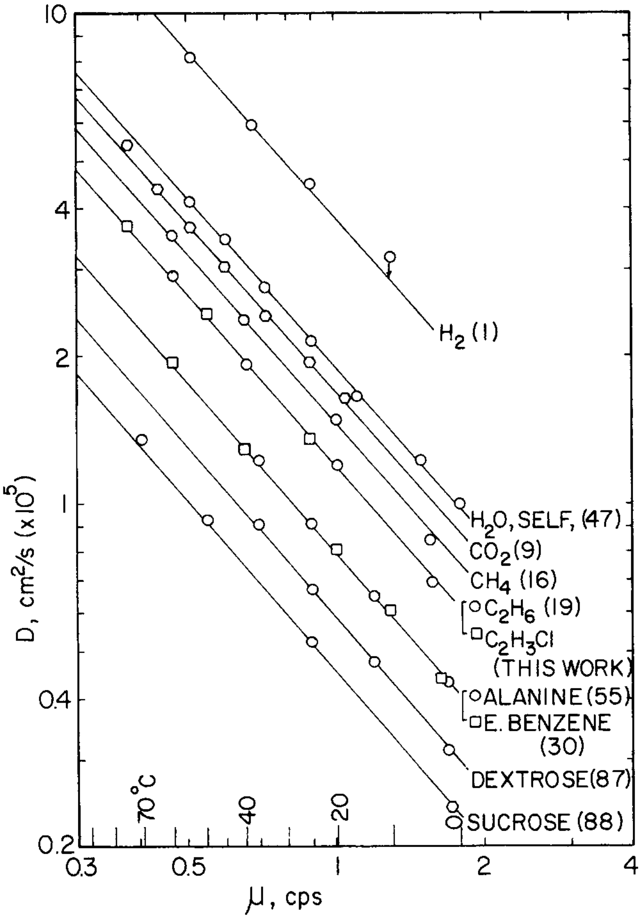

Fig. 1. Diffusivities in water as a function of solvent viscosity.

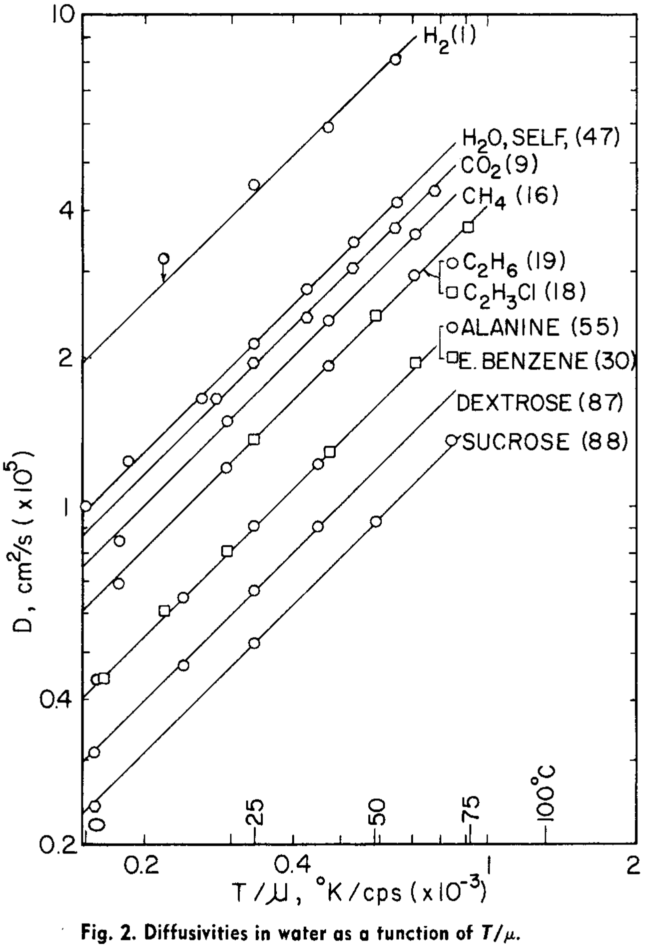

Fig. 2. Diffusivities in water as a function of T/μ.

in Table 3. It is noted that Equation (4) yields only slightly lower absolute errors than the original Othmer-Thakar equation and average errors which are actually slightly higher.

A number of conclusions can be made from the above analysis. Whereas the average error is somewhat reduced for all correlations except one by the use of true molar volumes, the average absolute error is similar for both molar volumes. It may be concluded that true molar volumes should be used if available but that the LeBas molar volumes would yield only slightly less accurate results. It is evident from Table 3 that Equation (4), the correlations of Othmer and Thakar, and of Wilke and Chang using $X = 2.26$, all yield nearly equivalent diffusivities in water, and that the Scheibel correlation yields diffusivities only 2 to 2.5% higher. This favorable comparison between the Othmer and Thakar and Scheibel correlations and that obtained in this work appears particularly meaningful because these comparisons are based on essentially new data, different from that on which the former two correlations were based. It may be concluded, therefore, that data published prior to 1950 are essentially as reliable as that available since 1950. In addition, it must be concluded that the Othmer-Thakar correlation is highly successful in its original form, in that the newly derived constants, as indicated in Equation (4), failed to significantly improve on the representation of the recent data. Finally, Figure 3 shows the relation between diffusivities predicted by the several correlations and the experimental data. In Figure 3 similar data for several substances are shown as a single point with the reference numbers referring to Table 2.

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NOTATION

D, D_{12} = diffusivity of dilute solute, cm²/s

$E_{avg} = \frac{1}{n} \sum^{n} \left[\frac{(D_{calc} - D)}{D} 100 \right]$, percent average error

$E_{abs} = \frac{1}{n} \sum^{n} \left[\frac{|D_{calc} - D|}{D} 100 \right]$, percent average absolute error

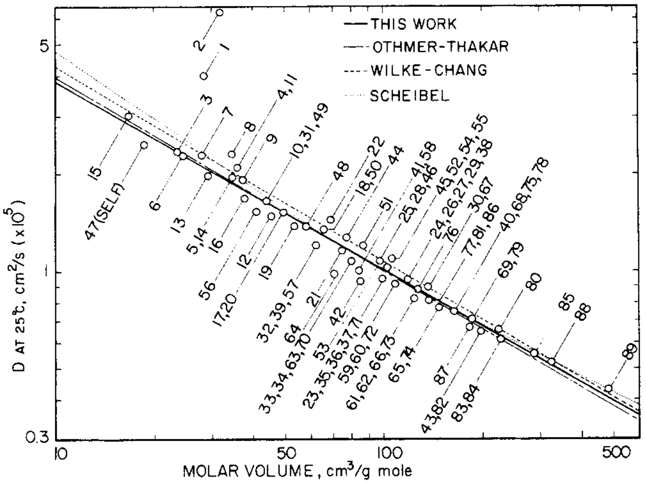

Fig. 3. Correlation of diffusivities in water as a function of solute molar volume at the normal boiling point.

$E_{max} = \left. \frac{(D_{calc} - D)}{D} 100 \right|_{max}$, maximum percent error

L = diffusion path length, cm

M_2 = molecular weight of solvent

n_A = mass flux of solute, g/cm^2 s

T = absolute temperature, °K

V_1, V_2 = molar volume at normal boiling point for solute (1), and solvent (2), cm^3/g mole

X = association parameter

x_2 = gas solubility at partial pressure of 1 atm., mole fraction

ρ = solution density, g/cm^3

μ, μ_2 = solution (solvent) viscosity, cps

ω_{A0} = mass fraction solute at gas-liquid interface

ω_{AL} = mass fraction solute at end of capillary

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