relation rules shows that the reaction

$$HN_3(^1A') \longrightarrow NH(^3\Sigma^-) + N_2(^1\Sigma_g^+)$$

giving products in the ground state is not allowed. However, reaction can proceed to the lowest singlet state of NH, thus

$$HN_3(^1A') \longrightarrow NH(^1\Delta) + N_2\,(^1\Sigma_g^+)$$

The $^1\Delta$ state is about 28 kcal./mole above the $^3\Sigma^-$ state of NH and thus would increase the activation energy for this decomposition to about 37 kcal./mole. A similar situation is discussed for the N_3 radical in a previous section. This activation barrier would render hydrazoic acid fairly stable at room temperature but would permit rapid decomposition above about 200°. We plan to study the kinetics of this reaction in an effort to determine this activation energy more definitely.

Consideration of the various tri-nitrogen entities shows that both N_3^- and N_3^+ are quite firmly bonded, but that N_3, if it exists at all, must be very unstable. Thus, $D(N_2\text{–}N^-)$ must be approximately equal to the electron affinity of N_3 and $D(N_2\text{–}N^+)$ is approximately 83 or 59 kcal./mole depending on whether N or N_2 retains the charge. On the other hand, apparently $D(N_2\text{–}N) \leqslant 0$. Presumably this is another example of the greater stability of systems having even numbers of electrons. It is striking that either adding or removing an electron permits formation of a firm bond where none otherwise exists.

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WASHINGTON, D. C.

[CONTRIBUTION FROM THE CHEMICAL LABORATORY OF NORTHWESTERN UNIVERSITY]

Diffusion in Sugar Solutions. IV. The Onsager Diffusion Coefficients for Glucose Diffusing in Sucrose Solutions

BY F. E. WEIR AND MALCOLM DOLE

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Height-area diffusion coefficients and reduced second moments have been measured at 25 and 35° for aqueous solutions containing on the average 60.5% sucrose, 0.5% glucose and 39% water. From these values the main and cross term diffusion coefficients as defined by Baldwin, Dunlop and Gosting have been calculated using the recently published method of Fujita and Gosting. Equations relating these diffusion coefficients to those defined by Onsager are given and the Onsager diffusion coefficients calculated.

I. Introduction

This paper represents the fourth in a series on diffusion in concentrated sucrose and glucose solutions and their mixtures. Having previously studied sucrose[1] and glucose[2] in binary systems with water as solvent and the diffusion of sucrose in the presence of glucose,[3] in this work the diffusion of glucose was studied in the presence of sucrose.

During the course of this work an important paper by Fujita and Gosting[4] appeared in which a mathematical technique was developed by which all four diffusion coefficients defined by the relations (first introduced by Baldwin, Dunlop and Gosting[5]), could be calculated. In eq. 1 the sub-

$$\frac{\partial c_1}{\partial t} = \mathfrak{D}_{11}\frac{\partial^2 c_1}{\partial x^2} + \mathfrak{D}_{12}\frac{\partial^2 c_2}{\partial x^2} \qquad (1a)$$

$$\frac{\partial c_2}{\partial t} = \mathfrak{D}_{22}\frac{\partial^2 c_2}{\partial x^2} + \mathfrak{D}_{21}\frac{\partial^2 c_1}{\partial x^2} \qquad (1b)$$

script 1 refers to sucrose and the subscript 2 to glucose. $\mathfrak{D}$ is the diffusion coefficient in cm.²/sec., x the distance from the initial boundary, c the solute concentration in moles/cc. and t the time.[6]

In this paper measured diffusion coefficients will be indicated by script $\mathfrak{D}$'s while coefficients calculated from the Onsager relations (see below) will be designated by Roman D's.

Experiments were conducted that made possible the use of Fujita–Gosting method. Experiments similar to those of Clarke and Dole[3] were also attempted, but with uncertain success. Included in the present paper are recalculated values of Clarke and Dole who inadvertently failed to add unity to the extrapolated j values to obtain j_m. (for definition see reference 1). The numerical values of Clarke and Dole were changed slightly by this correction.

II. Reciprocal Relations of Onsager

Some years ago, Onsager[7] generalized Fick's law of diffusion to systems of more than two components. For unidirectional diffusion in a system of 3 components

$$J_0 = -\mathrm{D}_{00}\frac{\partial c_0}{\partial x} - \mathrm{D}_{01}\frac{\partial c_1}{\partial x} - \mathrm{D}_{02}\frac{\partial c_2}{\partial x} \qquad (2a)$$

$$J_1 = -\mathrm{D}_{10}\frac{\partial c_0}{\partial x} - \mathrm{D}_{11}\frac{\partial c_1}{\partial x} - \mathrm{D}_{12}\frac{\partial c_2}{\partial x} \qquad (2b)$$

$$J_2 = -\mathrm{D}_{20}\frac{\partial c_0}{\partial x} - \mathrm{D}_{21}\frac{\partial c_1}{\partial x} - \mathrm{D}_{22}\frac{\partial c_2}{\partial x} \qquad (2c)$$

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[1] A. C. English and M. Dole, THIS JOURNAL, **72**, 3261 (1950).

[2] J. K. Gladden and M. Dole, *ibid.*, **75**, 3900 (1953).

[3] D. M. Clarke and M. Dole, *ibid.*, **76**, 3745 (1954).

[4] H. Fujita and L. J. Gosting, *ibid.*, **78**, 1099 (1956).

[5] R. L. Baldwin, P. J. Dunlop and L. J. Gosting, *ibid.*, **77**, 5235 (1955). See also P. J. Dunlop and L. J. Gosting, *ibid.*, **77**, 5238 (1955).

[6] It should be noted that the cross term coefficients $\mathfrak{D}_{12}$ and $\mathfrak{D}_{21}$ are different if the concentrations are expressed as grams/liter instead of moles/liter as first recognized by P. J. Dunlop. (Private communication from L. J. Gosting.) In this paper the concentrations will always be expressed in moles/liter or moles/cc.

[7] L. Onsager, *Ann. N. Y. Acad. Sci.*, **46**, 241 (1945). L. Onsager and R. M. Fuoss, *J. Phys. Chem.*, **36**, 2689 (1932). G. J. Hooyman. *Physica*, **22**, 751 (1956), has also discussed the Onsager reciprocal relations as applied to diffusion and has given this relation for ternary mixtures in terms of four diffusion coefficients.

where J represents the flux in units of moles/cm.^{2}/sec., and the subscript zero refers to the solvent water. These nine diffusion coefficients can be calculated from the measurable diffusion coefficients, $\mathfrak{D}_{11}$, $\mathfrak{D}_{12}$, $\mathfrak{D}_{22}$, $\mathfrak{D}_{21}$, by means of the following equations,[8] (concentration in moles/cc.)

$$D_{11} = \mathfrak{D}_{11}(1 - \bar{v}_1c_1) - \bar{v}_1c_2\mathfrak{D}_{12} \tag{3a}$$

$$D_{22} = \mathfrak{D}_{22}(1 - \bar{v}_2c_2) - \bar{v}_2c_1\mathfrak{D}_{21} \tag{3b}$$

$$D_{12} = \mathfrak{D}_{12}(1 - \bar{v}_2c_2) - \bar{v}_2c_1\mathfrak{D}_{11} \tag{3c}$$

$$D_{21} = \mathfrak{D}_{21}(1 - \bar{v}_1c_1) - \bar{v}_1c_2\mathfrak{D}_{22} \tag{3d}$$

$$D_{00} = (\mathfrak{D}_{11} + \mathfrak{D}_{22}) - (D_{11} + D_{22}) \tag{4a}$$

$$D_{10} = -\bar{v}_0(c_1\mathfrak{D}_{11} + c_2\mathfrak{D}_{12}) \tag{4b}$$

$$D_{20} = -\bar{v}_0(c_2\mathfrak{D}_{22} + c_1\mathfrak{D}_{21}) \tag{4c}$$

$$D_{01} = -\phi_1D_{11} - \phi_2D_{21} \tag{5a}$$

$$D_{02} = -\phi_1D_{12} - \phi_2D_{22} \tag{5b}$$

where the $\bar{v}$'s represent partial molal volumes in units of cc./mole, and ϕ_1 and ϕ_2 represent $\bar{v}_1/\bar{v}_0$ and $\bar{v}_2/\bar{v}_0$, respectively.

III. Experimental

The Gouy interference method, diffusion cell, optical equipment, other apparatus and materials were the same as those used in the previous work.[1-3] Considerable difficulty was experienced with water leaks between the glass plates of the diffusion cell and the water-bath, but after it was found that by using heavy grease ("Lubriseal") dissolved in a small amount of benzene to grease the faces of the stainless steel blocks, no more trouble occurred. The leaks were indicated by a large Δt (see below) and by an actual shift in the position of the initial boundary during the run.

Sucrose solutions approximately 60% by weight were prepared by dissolving special confectioner's sucrose in water at room temperature. Five ml. of 0.1 N KOH was added to the solution per 595 ml. of water in the solution. The refractive index of the solution was then determined with a dipping refractometer in order to find the concentration of the solutions so that weighed quantities of "special anhydrous dextrose" or sucrose could be added to bring the solution to the desired concentration by weight. The solutions were of three types: first those made according to the method of Clarke and Dole in which α_1 was kept approximately constant; second those made according to the requirements of the Fujita–Gosting method in which α_1 was varied from zero to unity while maintaining the average concentration of each component across the boundary approximately constant; and third, a solution of equal moles of sucrose and sucrose plus glucose on the two sides of the boundary which had been equilibrated in the vapor equilibrator used by Clarke and Dole[3] to equal vapor pressures of water. The equilibration was followed refractometrically until the refractive index showed no change with further equilibration. The definition of α_1 is given by the equation

$$\alpha_1 = \frac{\Delta n_1}{\Sigma \Delta n_i} = R_1 \frac{\Delta c_1}{\Sigma \Delta n_i} \tag{6}$$

where R_1 is the differential refractive index increment of sucrose at the average concentration of sucrose across the boundary. It was assumed that the total refractive index increment across the boundary, Δn, could be expressed by the equation

$$\Delta n = R_1\Delta c_1 + R_2\Delta c_2 \tag{7}$$

where c represents concentration in moles per liter. The concentration of each component was calculated from the weight percentages and the density. A few density measurements were made, but the density of all the solutions used was calculated taking the partial specific volumes of water, sucrose and glucose to be 0.9908, 0.6373 and 0.6439 cc./g. at 25°, respectively, and 0.9947, 0.6410 and 0.6472 cc./g. at 35°.

Equation 7 can be rearranged to

$$R_1 = \frac{\Delta n}{\Delta c_1} - \frac{\Delta c_2}{\Delta c_1} R_2 \tag{8}$$

so that it is easy to solve for R_1 and R_2 simultaneously by means of the least squares method. Equation 8 requires that $\Delta n/\Delta c_1$ be a linear function of $\Delta c_2/\Delta c_1$ provided that R_1 and R_2 are constant over the small concentration gradient of interest. The extent to which this was true in the experiments of this research is demonstrated in Fig. 1. Inasmuch as no difference could be detected between the 25 and 35° points on the curve, R_1 and R_2 were taken to be 0.04744 and 0.02500 liter/mole (uncertainty possibly several per cent.), respectively, at both temperatures.

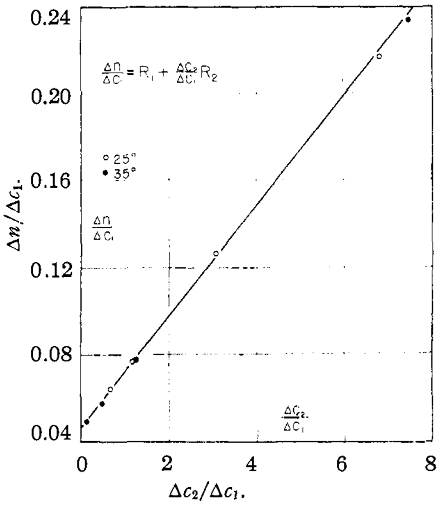

Fig. 1.—Test of eq. 8.

IV. Method of Calculating the Data

From the experimental observations of the shift in the Gouy fringes an average height–area ratio, $\mathfrak{D}_A$ can be calculated by the methods previously outlined by Gosting and Morris[9] and Akeley and Gosting.[10] If there is no interaction of flow, the individual diffusion coefficients of the two solutes, $\mathfrak{D}_{1A}$ and $\mathfrak{D}_{2A}$ are related to $\mathfrak{D}_A$ by the equation published by Gralen,[11]

$$\mathfrak{D}_A^{-1/2} = \alpha_1[\mathfrak{D}_{1A}^{-1/2} - \mathfrak{D}_{2A}^{-1/2}] + \mathfrak{D}_{2A}^{-1/2} \tag{9}$$

The use of this equation requires that the average sucrose and glucose concentrations remain constant as α_1 is varied. By plotting $\mathfrak{D}_A^{-1/2}$ as a function of α_1, $\mathfrak{D}_{1A}$ and $\mathfrak{D}_{2A}$ may be calculated from appropriate relations involving the slope and intercept providing that the data follow the required linear relationship. Figure 2 illustrates a plot of $\mathfrak{D}_A^{-1/2}$ as a function of α_1 for 25 and 35°. Satisfactory agreement with the required linear relationship is seen to exist. In the work of Clarke and Dole in which α_1 was held constant, $\mathfrak{D}_{1A}$ was calculated from eq. 9 assuming $\mathfrak{D}_{2A}$ to be the same as $\mathfrak{D}_2$ for the pure glucose–water system (in that work glucose solutions were the solvent). The results of the present work discussed below show that this assumption was reasonable.

Another approach to the interpretation of the $\mathfrak{D}_A$ values is to follow the method of Fujita and Gosting[4] in calculating $\mathfrak{D}_{11}$, $\mathfrak{D}_{22}$, $\mathfrak{D}_{12}$, and $\mathfrak{D}_{21}$, in which the reduced second moment, $\mathfrak{D}_{2m}$, is evaluated by a rather complicated mathematical and graphical method (see the paper of Fujita and Gosting for details).

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[8] M. Dole, *J. Chem. Phys.*, **25**, 1082 (1956).

[9] L. J. Gosting and M. S. Morris, THIS JOURNAL, **71**, 1998 (1949).

[10] D. F. Akeley and L. J. Gosting, *ibid.*, **75**, 5685 (1953).

[11] N. Gralen, *Kolloid Z.*, **95**, 188 (1941); O. Quensel, Dissertation, Uppsala, 1942.

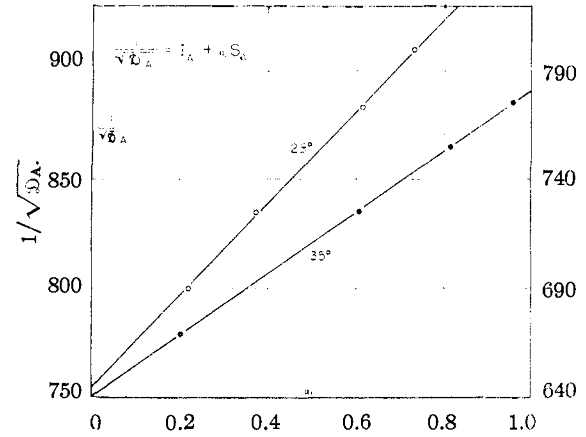

Fig. 2.—Test of eq. 12 and 17.

Figure 3 illustrates a plot of $\mathfrak{D}_{2m}$ at 25 and 35° as a function of α_1 which according to the theory of Fujita and Gosting should result in a straight line inasmuch as

$$\mathfrak{D}_{2m} = I_{2m} + S_{2m}\alpha_1 \qquad (10)$$

where I_{2m} and S_{2m}, the intercept and slope, respectively, of the line, are related to $\mathfrak{D}_{22}$, $\mathfrak{D}_{11}$, $\mathfrak{D}_{12}$ by equations given by Fujita and Gosting.[4]

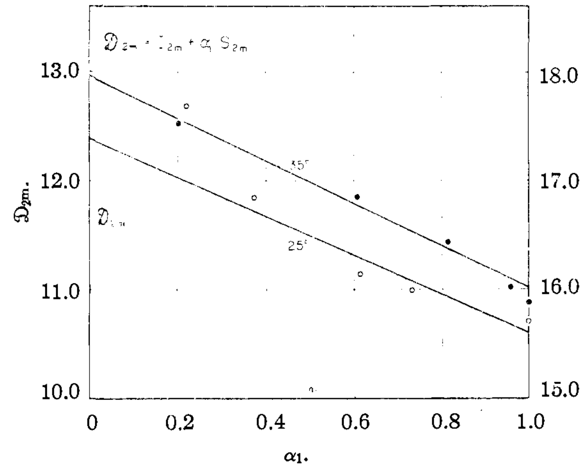

Fig. 3.—Test of eq. 16.

The value of $\mathfrak{D}_{2A}$ for the water vapor equilibrated solution was calculated from eq. 9 making use of the assumption of Clarke and Dole that $\mathfrak{D}_{1A}$ is equal to $\mathfrak{D}_1$ in such a system.

V. Experimental Results

All of the experimental results are collected in Tables I and II. It is difficult to estimate the reliability of the data, especially the diffusion coefficients calculated by the Fujita–Gosting method. This is because two graphical integrations have to be carried out and because in the calculations the diffusion coefficients depend upon small differences between large numbers. The critical calculation is that of $\mathfrak{D}_{2m}$. In one experiment at 35°, one of us obtained 15.95×10^{-7} for this value while the other found 16.86×10^{-7} (using the same fringe deviation values); in another experiment the values were 15.79×10^{-7} and 16.44×10^{-7}, respectively. The values plotted in Fig. 3 and used in the calculations were those calculated by the senior author. Looking at Fig. 3 it can be seen that the 25° value of $\mathfrak{D}_{2m}$ at α_1, equal to 0.219 is considerably out of line with the other data at that temperature. The Δt value given in Table II is also inordinately high. In computing the constants of the straight line for 25° we have omitted this value of $\mathfrak{D}_{2m}$ and have substituted for it the value of $\mathfrak{D}_A$ calculated for α_1 equal to unity, assuming thereby that at this value of α_1 there will be no fringe deviations. The fringe deviations plotted in Fig. 4 for two values of α_1 suggest that at α_1 equal to unity the fringe deviations might be zero. In the

TABLE I

REDUCED HEIGHT–AREA RATIOS AS CALCULATED FROM EQUATION 12

All diffusion coefficients in cm.² sec. × 10⁷. Concentration in moles/liter of solution.

	25°			
Exp. no.	11	13	17	35 (Water equilibrated)
Av. c_1	2.232	2.218	2.218	2.255
Av. c_2	0.07153	0.1436	0.1800	0.07144
Δc_1	− .0373	− .0751	− .0946	.1089
Δc_2	.1431	.2872	.3600	− .1429
α_1	−1.040	−1.054	−1.062	+ 3.384
Δt, sec.	20	213	341	−54
$\mathfrak{D}_A$	15.69	14.05	12.20	3.504
$\mathfrak{D}_1$[a]	11.60	11.88	11.92	11.30
$\mathfrak{D}_{2A}$	13.38	12.87	12.05	25.47

Fujita–Gosting type experiment (see also Table II)

	$\mathfrak{D}_{1A}$	$\mathfrak{D}_{2A}$
25°	10.73	17.57
35°	16.39	24.39

[a] Calculated from data of English and Dole at the average sucrose concentration shown above.

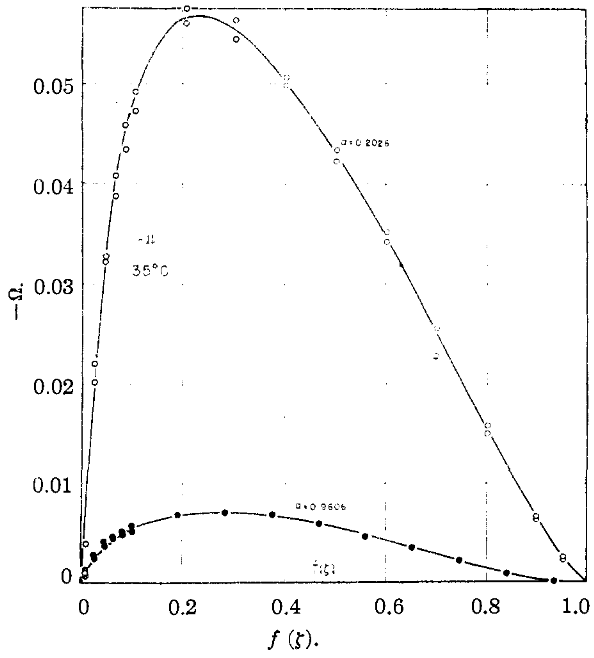

Fig. 4.—Interference fringe deviation from Gaussian behavior at two values of α_1.

TABLE II

DATA FOR DIFFUSION COEFFICIENTS CALCULATED BY THE FUJITA–GOSTING METHOD

All diffusion coefficients × 10⁷ in cm.²/sec. Concentrations in moles/liter of solutions.

	25°				35°			
Expt. no.	36	34	24	25	37	42	39	40
Av. c_1	2.277	2.275	2.278	2.280	2.266	2.272	2.269	2.269
Av. c_2	0.03231	0.03733	0.03596	0.03586	0.03569	0.03576	0.3550	0.03642
Δc_1	.0094	.0245	.0621	.0532	.0096	.0498	.0385	.0487
Δc_2	.0646	.0746	.0719	.0360	.0714	.06152	.01769	.00517
α_1	.2186	.3754	.6175	.7363	.2026	.6102	.8172	.9606
Δt, sec.	830	68	−41	−25	60	156	51	20
$\mathfrak{D}_A$	15.63	14.34	12.81	12.07	22.27	19.07	17.60	16.56
$\mathfrak{D}_{2m}$	12.69	11.84	11.14	11.00	17.53	16.86	16.44	16.03

	$\mathfrak{D}_{11}$	$\mathfrak{D}_{22}$	$\mathfrak{D}_{12}$	$\mathfrak{D}_{21}$
25°	10.95	2.37	5.27	−0.64
35°	17.16	2.18	8.32	−2.13

case of the $\mathfrak{D}_{2m}$ values at 35° the straight line extrapolated to α_1 equal to unity yields a value of $\mathfrak{D}_{2m}$ not much greater than the value of $\mathfrak{D}_A$ calculated for α_1 equal to unity. It is believed that this procedure yielded more reliable values for I_{2m} and S_{2m} at 25° than would have been obtained if the $\mathfrak{D}_{2m}$ value at α_1 equal to 0.219 had been included in the least squares calculation. The straight lines of Figs. 2 and 3 illustrate the values calculated by the least squares equation.

Table III contains values of the Onsager diffusion coefficients calculated by means of eq. 3a to 5b, inclusive.

TABLE III

DATA FOR THE ONSAGER DIFFUSION COEFFICIENTS IN CM.²/SEC. × 10⁷

	25°	35°
	61% Sucrose—two components[1]	
D_{00}	5.34	7.98
D_{01}	−64.8	−96.7
D_{10}	− 0.44	− 0.65
D_{11}	5.36	7.90
	Three components 60.5% sucrose, 0.5% glucose, 39% water av. concn.	
D_{00}	5.33	8.05
D_{01}	−64.5	−97.6
D_{02}	−45.2	−64.0
D_{10}	− 0.45	− 0.71
D_{11}	5.46	8.58
D_{12}	2.36	3.79
D_{20}	0.025	0.09
D_{21}	− 0.34	− 1.14
D_{22}	2.53	2.71

In the introduction it was stated that Clarke and Dole failed to add unity to their estimates of j_m. Table IV contains a summary of Clarke and Dole's data as recalculated using the new values of j_m.

VI. Discussion of Data

It is first of all of interest to compare the $\mathfrak{D}_{1A}$ and the $\mathfrak{D}_{2A}$ with the $\mathfrak{D}_1$ and $\mathfrak{D}_2$ diffusion coefficients measured by English and Dole[1] and Gladden and Dole[2] in the binary systems. Such comparison for the component present in small concentration, glucose in this work, is questionable because of the change in the nature of the solvent and because there is uncertainty as to whether the comparison should be made at constant mole fraction of combined sugars or constant weight fraction. There should be less uncertainty in comparing diffusion coefficients for the sucrose which was present at 60.5 wt. % concentration. At 25° and 61.05% sucrose English and Dole[1] found 10.71 × 10^{-7} cm.²/sec., while 10.73 × 10^{-7} also was calculated from experiments 36, 34, 24 and 25 of this research.

TABLE IV

CORRECTED REDUCED HEIGHT–AREA RATIOS OF CLARKE AND DOLE COEFFICIENTS IN CM.²/SEC. × 10⁷

Av. wt. % Glucose	Sucrose	A
	25°	
5	0	
5.176	0.999	47.92
3.00	0	
29.82	0.600	26.44
29.73	0.900	26.13
29.55	1.500	25.83
4.00	0	
39.93	1.501	11.56
50.0	0	
49.70	0.601	10.94
49.50	1.000	11.51
49.37	1.467	11.18
49.23	1.540	11.36
49.06	2.393	11.21
60	0	
59.48	0.953	5.192
59.77	1.520	4.774
58.95	1.998	5.047
58.29	2.516	5.099
70.0	0	
70.40	0.966	1.270
69.99	1.485	1.328
	35°	
60.0	0	
59.47	1.004	8.567
59.51	1.017	8.254
59.30	1.512	7.966

At 35° the agreement was not as good, 15.88 × 10^{-7} found by English and Dole again at 61.05 % sucrose, and 16.39 × 10^{-7} from this research. These results seem to be good enough to justify the assumption made by Clarke and Dole[3] that $\mathfrak{D}_{1A}$ of eq. 9 is approximately equal to $\mathfrak{D}_1$ at the same total weight fraction of sugar if one of the components is present in small percentage.

Clarke and Dole found that $\mathfrak{D}_{1A}$ for the diffusion of sucrose in glucose solutions as solvent was greater than $\mathfrak{D}_1$ if the comparison was made at the same total mole fraction of solute. In this research it was found as expected that $\mathfrak{D}_{2A}$ as calculated from eq. 9 at α_1 equal to zero, 17.57×10^{-7} cm.^{2}/sec., was smaller than $\mathfrak{D}_2$ for glucose, 25.7×10^{-7} calculated from the data of Gladden and Dole[2] at the same total mole fraction of solute. Thus, sucrose diffuses faster in solutions of glucose than in its own solutions while glucose diffuses more slowly in solutions of sucrose than in its own solutions. If, however, the comparison is made at the same weight fraction, the converse is true, as was also found by Clarke and Dole.[3] Assuming no interaction of solute flows, the diffusion coefficient of glucose, $\mathfrak{D}_{2A}$ can be calculated using eq. 9 from the three types of experiments done in this work, namely, from the Fujita–Gosting, "opposed gradients" and water equilibrated experiments. The results are, in order mentioned at 25°, 17.57, 14.2 (value extrapolated to zero concentration of glucose) and 25.47×10^{-7} cm.^{2}/sec. In the Fujita–Gosting type experiments the concentration gradients for sucrose and glucose were parallel, *i.e.*, they had the same sign, whereas in the Clarke–Dole type the concentration gradients had the opposite sign. In the latter the diffusion of sucrose was in the opposite direction or "opposed" the diffusion of glucose. The α-values were approximately -1.0 for the opposed gradient experiments,[1] $+0.5$ (average) for the parallel gradient and $+3.38$ for the water equilibrated experiment. Thus there is a very good correlation between the calculated $\mathfrak{D}_{2A}$ values and α_1, the larger the sucrose gradient, the larger is the calculated diffusion coefficient of glucose. In fact a plot of $\mathfrak{D}_{2A}$ for the three types of experiments as a function of α_1 approximates well to a straight line. The value of $\mathfrak{D}_A$ for the water equilibrated experiment at α_1 equal to 3.38 does not "fit" on the extrapolated straight line of Fig. 2, nor do the $\mathfrak{D}_A$ values of the "opposed gradient" type agree at all with the relationship of Fig. 2. In fact, approximately the same values of $\mathfrak{D}_A$ were obtained as in the Fujita–Gosting experiments but at negative α-values.

At this point it should be noted that many of the experiments of the Clarke–Dole type which were attempted failed to give reliable results. The fringe photographs were imperfect, the fringes on the photographic plate showed a variable density from top to bottom and sometimes an apparent splitting of a fringe into two fringes appeared. In experiments no. 11, 13 and 17 of Table I. C_t calculated from the larger fringe distances did not change with fringe number as expected. For fringe numbers above 10, non-Gaussian behavior existed. The variation of the fringe deviation Ω with time should be constant, but this was not the case as the peculiar result of zero values of Ω for fringes up to 10 did not appear until diffusion had proceeded for about 5000 seconds. A clear explanation of these observations is not available at the present time. All of these experiments were performed before the best technique of assembling the cell was developed. The data are included in Table I, nevertheless, because other criteria for valid experimental results such as not too large Δt values and fairly good agreement between the $\mathfrak{D}_{2A}$ values indicated that the results were possibly significant.

In the mathematical analysis of Gosting and his school, an attempt has been made to develop theoretical techniques for determining the extent of interaction between different diffusing species. If there were no interaction of flows, then the $\mathfrak{D}_{12}$ and $\mathfrak{D}_{21}$ diffusion coefficients should be zero. From Tables II and III it is clear that interaction did occur. Referring to Table III one sees that $\mathfrak{D}_{00}$ and D_{11} are practically the same in the three component solution as in the two component as might be expected as the small amount of glucose present should not greatly affect the diffusion of sucrose or water due to their own concentration gradients.

A surprising result is the low value of D_{22}, about one tenth that calculated assuming no interaction of flows. From a physical standpoint such a low result would have to be explained on the basis, perhaps, of complex formation between sucrose and glucose molecules.

Comparing the D_{21} with the D_{12} values the former is very small and negative, while D_{12} is quite large. In other words the diffusion coefficient representing the flow of glucose due to the sucrose concentration gradient is not as great as that for the flow of sucrose due to the glucose concentration gradient.

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