*J. Chem. Eng. Data* **2007,** *52,* 40−43

# Infinite Dilution Binary Diffusion Coefficients for Six Sugars at 0.1 MPa and Temperatures from (273.2 to 353.2) K

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Infinite dilution binary diffusion coefficients $D_{12}$ for arabinose, xylose, glucose, mannose, galactose, and sucrose in water were measured over the temperature range from (273.2 to 353.2) K at 0.1 MPa with the Taylor dispersion method. The values of $D_{12}/T$ were well correlated with water viscosity for each solute with average absolute deviation AAD < 1.7 %. The accuracies for various predictive correlations were also examined.

## Introduction

Diffusion coefficients for sugars in water are of importance for reactor design in purification or isolation processes such as chromatography, membrane filtration, or crystallization in food, neutraceutical, and pharmaceutical industries. Measurements of binary diffusion coefficients have been reported by various methods, and most data were measured by interferometric techniques such as the Gouy,[1−7] the Rayleigh,[5,8−11] the Schlieren,[2] the Mach−Zehnder,[12] and the Jamin methods[13] as well as other techniques such as the diaphragm cell,[14,15] the Stephan capillary,[16] and the Taylor dispersion method.[17] Yamamoto and Sano[18] also determined binary diffusion coefficients for various sugars at 303 K from drying rates. Although a large number of diffusion coefficient data for sugars in water or aqueous solutions have been reported, the data are available mainly for sucrose[1−6,8,9,12−16,18] and glucose,[3,7−9,17,18] in particular, at ambient temperature. The accuracy of the Taylor dispersion method is relatively high[19] and adequate for measuring binary diffusion coefficients over a wide temperature range. In this study, infinite dilution binary diffusion coefficients $D_{12}$ for six sugars in water were measured by the Taylor dispersion method at atmospheric pressure over the temperature range from (273.2 to 353.2) K by fitting the calculated response curve to that measured experimentally. Accuracies of various correlations for predicting the $D_{12}$ values were also examined.

## Experimental Section

The experimental apparatus and procedures are almost the same as those in the previous measurements for $C_5$-alcohols in water,[20] but a syringe pump (DM100, ISCO) and a differential refractive index detector (model L-7490, Hitachi, Japan) were used. Distilled water was degassed and supplied by the syringe pump to a fused silica diffusion tubing with 0.5274 mm diameter, 0.25 m coil diameter, and 31.17 m long. A sugar sample as an aqueous solution in mass fraction of 0.001 was injected through a HPLC sample injector (Rheodyne 7250) with a sampling loop of 20 $\mu$L. The detector linearity, such that the signal intensity is proportional to the sugar concentration, was confirmed by injecting each sugar solution at different concentrations. D-Arabinose (99 %, Aldrich), D-xylose (99 %, Aldrich), D-glucose (99.5 %, Sigma), D-mannose (99 %, Aldrich), D-galactose (99 %, Sigma), and sucrose (99+ %, Aldrich) were used without further purification.

**Table 1. Comparison between the Present Study and Literature Data for Xylose, Glucose, and Sucrose at 298 K**

| source | method | 100 m[a] | $10^9 D_{12}$/ $m^2 \cdot s^{-1}$ |
|---|---|---|---|
| | Xylose | | |
| present study | Taylor | 0 | 0.769 |
| ref 10 | Rayleigh | 0 | 0.7495[a] |
| | Glucose | | |
| present study | Taylor | 0 | 0.676 |
| ref 3 | Gouy | 0 | 0.675[b] |
| ref 9 | Rayleigh | 0.78 | 0.6728 |
| ref 7 | Gouy | 0 | 0.673[b] |
| ref 17 | Taylor | 0 | 0.69 |
| | Sucrose | | |
| present study | Taylor | 0 | 0.546 |
| ref 1 | Gouy | 0 | 0.5226[b] |
| ref 2 | Gouy | 0.73 | 0.5174 |
| ref 3 | Gouy | 0 | 0.523[b] |
| ref 9 | Rayleigh | 0.77 | 0.5209 |
| ref 4 | Gouy | 0 | 0.5228[b] |
| ref 5 | Gouy | 0 | 0.5178[b] |
| ref 12 | Mach−Zehnder | 0.60 | 0.5212 |
| ref 14 | diaphragm cell | 0 | 0.523[b] |
| ref 13 | Jamin | 0.1 | 0.5219 |
| ref 15 | diaphragm cell | 0.0007 | 0.524 |
| ref 6 | Gouy | 0.70 | 0.518 |

[a] m, mass fraction. [b] Extrapolated value.

The effect of the secondary flow due to the diffusion column coiling is negligible because the criterion, $DeSc^{1/2} < 8$ , is fulfilled for all measurement conditions, where *De* is the Dean number $De = (\rho u d_{\mathrm{tube}}/\eta)(d_{\mathrm{tube}}/d_{\mathrm{coil}})^{1/2}$; $d_{\mathrm{tube}}$ and $d_{\mathrm{coil}}$ are the inner diameter of diffusion column and the coil diameter, respectively; and the Schmidt number $Sc = \eta/\rho D_{12}$; $\rho$ and $\eta$ are the water density and viscosity, respectively. Alizadeh et al.[21] estimated the error caused by the secondary flow due to column coiling as a function of $De^2Sc$. When the criterion is satisfied, the error in terms of the second moment is less than 1 %. In most runs the fitting error $\epsilon$ was about 0.01, defined by eq 3. The corresponding uncertainty for the $D_{12}$ values is within ± 3 %, mainly ± 2.5 %. The uncertainty of temperature measured is ± 0.1 K at temperatures up to 333 K and ± 0.2 K above this temperature.

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10.1021/je0601816 CCC: $37.00

**Table 2. Measured Binary Diffusion Coefficients of Sugars in Water at 0.1 MPa**

| compound | formula | $T$/K | $D_{12}/10^{-9}$ m$^2$·s$^{-1}$ | $u/10^{-2}$ m·s$^{-1}$ | $10^2\,\epsilon$ | $De\cdot Sc^{1/2}$ |
|---|---|---|---|---|---|---|
| arabinose | $C_5H_{10}O_5$ | 273.2 | 0.359 | 0.528−0.529 | 1.10−1.43 | 5.05−5.06 |
| | | 293.2 | 0.682 | 0.530 | 0.62−0.94 | 4.91 |
| | | 298.2 | 0.773 | 0.529−0.530 | 0.64−1.21 | 4.88−4.89 |
| | | 313.2 | 1.11 | 0.532 | 0.97−1.31 | 4.77 |
| | | 333.2 | 1.67 | 0.537−0.538 | 0.93−1.40 | 4.62−4.63 |
| | | 353.2 | 2.37 | 0.544−0.546 | 0.70−0.77 | 4.48−4.50 |
| xylose | $C_5H_{10}O_5$ | 273.2 | 0.348 | 0.529−0.530 | 0.86−1.07 | 5.14−5.15 |
| | | 293.2 | 0.665 | 0.529−0.530 | 0.81−1.58 | 4.96−4.97 |
| | | 298.2 | 0.769 | 0.529−0.530 | 0.81−1.21 | 4.89−4.90 |
| | | 313.2 | 1.11 | 0.531−0.535 | 0.72−1.03 | 4.81−4.83 |
| | | 333.2 | 1.64 | 0.537−0.539 | 0.71−1.14 | 4.66−4.68 |
| | | 353.2 | 2.33 | 0.542−0.544 | 0.62−1.00 | 4.50−4.52 |
| glucose | $C_6H_{12}O_6$ | 273.2 | 0.323 | 0.529−0.530 | 1.18−1.55 | 5.33−5.34 |
| | | 293.2 | 0.605 | 0.529−0.531 | 0.47−1.42 | 5.20−5.22 |
| | | 298.2 | 0.676 | 0.526−0.530 | 0.98−1.66 | 5.19−5.23 |
| | | 313.2 | 0.997 | 0.533 | 0.58−0.92 | 5.04 |
| | | 333.2 | 1.49 | 0.537−0.539 | 0.59−1.29 | 4.89−4.91 |
| | | 353.2 | 2.13 | 0.543 | 0.64−0.87 | 4.72 |
| mannose | $C_6H_{12}O_6$ | 273.2 | 0.334 | 0.529−0.530 | 0.89−1.52 | 5.24−5.25 |
| | | 293.2 | 0.620 | 0.530−0.531 | 0.81−1.51 | 5.15−5.16 |
| | | 298.2 | 0.704 | 0.529−0.531 | 0.85−1.36 | 5.11−5.13 |
| | | 313.2 | 1.02 | 0.532−0.533 | 0.72−0.96 | 4.97−4.98 |
| | | 333.2 | 1.59 | 0.537−0.538 | 0.99−1.40 | 4.74 |
| | | 353.2 | 2.16 | 0.543−0.544 | 0.54−0.69 | 4.68−4.69 |
| galactose | $C_6H_{12}O_6$ | 273.2 | 0.319 | 0.529−0.530 | 0.773−1.43 | 5.37−5.38 |
| | | 293.2 | 0.625 | 0.529−0.530 | 0.81−1.13 | 5.12−5.13 |
| | | 298.2 | 0.706 | 0.530−0.531 | 0.76−1.08 | 5.11−5.12 |
| | | 313.2 | 1.01 | 0.532−0.533 | 0.70−1.15 | 5.00−5.01 |
| | | 333.2 | 1.51 | 0.538 | 0.69−1.31 | 4.87 |
| | | 353.2 | 2.17 | 0.544−0.545 | 0.54−0.60 | 4.68−4.69 |
| sucrose | $C_{12}H_{22}O_{11}$ | 273.2 | 0.255 | 0.529−0.530 | 1.24−2.04 | 6.00−6.01 |
| | | 293.2 | 0.487 | 0.529 | 1.01−1.49 | 5.80 |
| | | 298.2 | 0.546 | 0.529−0.530 | 0.70−1.08 | 5.80−5.81 |
| | | 313.2 | 0.792 | 0.531−0.537 | 0.96−1.63 | 5.63−5.70 |
| | | 333.2 | 1.17 | 0.537−0.539 | 0.93−0.16 | 5.52−5.54 |
| | | 353.2 | 1.69 | 0.540−0.546 | 0.46−0.74 | 5.27−5.31 |

## Analysis

When a dilute aqueous sugar solution was injected at $z = 0$ to a fully developed laminar flow moving in a circular cross-sectional diffusion tubing, the sugar concentration at column exit $z = L$ can be expressed by[22,23]

$$c(t) = \frac{m}{\pi R_0^2 \sqrt{4\pi K t}} \exp\left[-\frac{(L - ut)^2}{4Kt}\right] \tag{1}$$

where

$$K = D_{12} + \frac{R_0^2 u^2}{48 D_{12}} \tag{2}$$

and $c(t)$ is the solute concentration, $m$ is the amount of the sugar injected, $u$ is the average velocity of water, $R_0$ is the tube inner radius of the diffusion column, and $D_{12}$ is the binary diffusion coefficient. In this study, two parameters of $D_{12}$ and $u$ were determined so that the root-mean-square fitting error $\epsilon$ defined by eq 3 is minimized:

$$\epsilon = [\int_{t_1}^{t_2} (C_{\mathrm{cal}}(t) - C_{\mathrm{exp}}(t))^2\,\mathrm{d}t / \int_{t_1}^{t_2} (C_{\mathrm{exp}}(t))^2\,\mathrm{d}t]^{1/2} \tag{3}$$

where $C_{\mathrm{cal}}(t)$ and $C_{\mathrm{exp}}(t)$ are the calculated and experimentally measured concentrations, respectively, both normalized by peak area ($C(t) = c(t)/\int_0^\infty c(t)\,\mathrm{d}t$), and $t_1$ and $t_2$ ($t_1 < t_2$) are the times that correspond to the 10 % peak height of the response curve.[24,25] The integration in eq 3 was numerically made by the Simpson's 1/3 rule method at time intervals $\Delta t = (0.01$ to $0.1)$ min (mainly 0.1 min). In practice, the $\epsilon$ values were calculated for various sets of assumed $D_{12}$ and $u$ values, and the best fit values of $D_{12}$ and $u$ with the minimum $\epsilon$ value were determined by depicting an error contour map of $D_{12}$ versus $u$ values.[24,25]

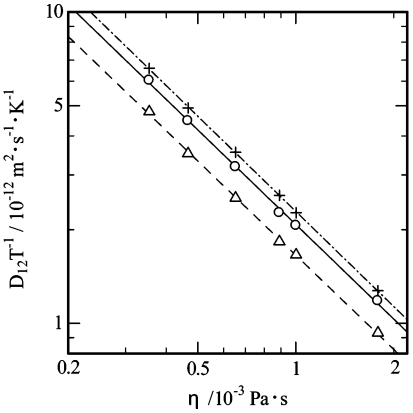

**Figure 1.** $D_{12}/T$ vs water viscosity for sugars at temperatures from (273.2 to 353.2) K: +, xylose; ○, glucose; △, sucrose.

## Results and Discussion

Table 1 compares $D_{12}$ values measured in the present study and those reported in the literature for xylose, glucose, and sucrose at 298 K. The present datum for glucose is almost in agreement with the literature data while the present value for

**Table 3. Values of Constants Involved in Equation 4**

| solute[a] | arabinose | xylose | glucose | mannose | galactose | sucrose |
|---|---|---|---|---|---|---|
| $10^{15}\,\alpha$ | 2.224 | 2.057 | 1.947 | 2.011 | 1.855 | 1.602 |
| $\beta$ | −1.007 | −1.016 | −1.009 | −1.010 | −1.019 | −1.005 |
| 100AAD | 0.81 | 0.52 | 1.38 | 1.70 | 0.86 | 1.17 |

[a] Units of α and $\beta$ are based on $D_{12}$/m$^2$·s$^{-1}$ and $\eta$/Pa·s.

**Table 4. Accuracies for Various Predictive Correlations**

| | 100ADD | | | | | | |
|---|---|---|---|---|---|---|---|
| correlation | arabinose | xylose | glucose | mannose | galactose | sucrose | overall |
| Wilke and Chang[29] | | | | | | | |
| $\varphi = 2.6$ | 13.5 | 15.3 | 14.6 | 10.8 | 12.5 | 3.0 | 11.6 |
| $\varphi = 2.26$[30] | 5.8 | 7.5 | 6.8 | 3.3 | 4.9 | 9.6 | 6.3 |
| Hayduk and Laudie[30] | 5.6 | 7.3 | 6.8 | 3.7 | 5.1 | 8.9 | 6.3 |
| Tyn and Calus[29] | 30.6 | 31.7 | 33.4 | 31.1 | 32.2 | 30.5 | 31.6 |
| Nakanishi[29] | 1.1 | 1.0 | 1.9 | 2.0 | 1.0 | 6.5 | 2.3 |
| Hayduk and Minhas[29] | 9.5 | 10.2 | 8.8 | 6.4 | 6.8 | 33.4 | 12.5 |
| Siddiqi and Lucas[31] | 9.4 | 10.1 | 8.7 | 6.3 | 6.7 | 33.4 | 12.4 |
| eq 5 | 2.7 | 3.4 | 2.2 | 2.8 | 3.4 | 2.9 | 2.9 |
| $10^6 A/\mathrm{m^2{\cdot}s^{-1}}$ | 1.464 | 1.511 | 1.334 | 1.377 | 1.423 | 1.026 | |
| $E/\mathrm{kJ{\cdot}mol^{-1}}$ | 18.77 | 18.89 | 18.83 | 18.82 | 18.95 | 18.74 | |

sucrose ($0.546 \times 10^{-9}$ m$^2$·s$^{-1}$) is higher than the literature values by 4.8 % from the mean value of $0.521 \times 10^{-9}$ m$^2$·s$^{-1}$ of the data measured by the optical methods.[1−6,9,12−14] The reason is not clear, but $D_{12}$ for glucose by van de Ven-Lucassen and Kerkhof[19] with the Taylor dispersion method is also somewhat higher. Table 2 lists the measured $D_{12}$ values, which are the mean values from three or four measurements at each condition, together with the ranges of $u$, $\epsilon$, and $De{\cdot}Sc^{1/2}$.

Figure 1 shows logarithmic plots of $D_{12}/T$ versus water viscosity for xylose, glucose, and sucrose at temperatures from (273.2 to 353.2) K. The $D_{12}$ values for each solute were well-represented with each straight line in eq 4:

$$D_{12}/\mathrm{m^2{\cdot}s^{-1}} = \alpha(\eta/\mathrm{Pa{\cdot}s})^{\beta}(T/\mathrm{K}) \qquad (4)$$

where $D_{12}$ is the binary diffusion coefficient, $\alpha$ and $\beta$ are constants, $T$ is the temperature, and $\eta$ is the water viscosity. Equation 4 is effective, while constants $\alpha$ and $\beta$ are specific to each solvent−solute system for $D_{12}$ values for C$_5$-monoalcohol in water[20] and for organic compounds in supercritical CO$_2$,[26] in mixtures of hexane and dense CO$_2$,[27] and in liquid organic solvents.[28] Table 3 presents the values of $\alpha$ and $\beta$ determined for each solute, and the average absolute deviations (AAD), defined as $(1/N)|(D_{12,\mathrm{prd}}/D_{12,\mathrm{exp}}) - 1|$, where $N$ is the number of data points. $D_{12,\mathrm{prd}}$ and $D_{12,\mathrm{exp}}$ are the predicted and experimentally measured values. If the Stokes−Einstein equation can be applied, the $\beta$ value should be $-1.0$. The values of $\beta$ range from $-1.019$ to $-1.005$. Similarly, the absolute values of $\beta$ for C$_5$-monoalcohols in water are slightly higher than unity.[20]

Table 4 lists the AAD values for various correlations tested for the present $D_{12}$ values. Although the most correlations listed in Table 4 are based on the Stokes−Einstein equation, the Arrhenius-type equation given in eq 5 is also examined:

$$D_{12}/\mathrm{m^2{\cdot}s^{-1}} = (A/\mathrm{m^2{\cdot}s^{-1}}) \exp\left[-\frac{E/\mathrm{J{\cdot}mol^{-1}}}{(R/\mathrm{J{\cdot}K^{-1}{\cdot}mol^{-1}})(T/\mathrm{K})}\right] \qquad (5)$$

where $A$, $E$, and $R$ are the pre-exponential factor, the activation energy, and the gas constant, respectively. Hayduk and Minhas[29] modified the association factor $\varphi$ in the Wilke and Chang equation to be 2.26 instead of 2.6 in the original equation. Nakanishi's equation,[29] when the solute interaction parameter $I_1 = 1.5$, is most accurate while the accuracy for sucrose is lower. Equation 5 is also preferable, and the Hayduk and Laudie[30] equation and the Wilke and Chang equation with $\varphi = 2.26$ are acceptable. The Hayduk and Minhas, the original Wilke and Chang[29] with $\varphi = 2.6$, the Tyn and Calus,[29] and the Siddiqi and Lucas equations[31] are less accurate. As compared with these equations, the correlation with viscosity in eq 4 is found to be more accurate while the two adjustable constants $\alpha$ and $\beta$ are involved: these values are specific to solutes. The accuracy in the Arrhenius-type equation in eq 5, which has also two adjustable constants for each solute, is better than those based on the Stokes−Einstein relationship. However, the accuracy in eq 5 is lower than that in eq 4.

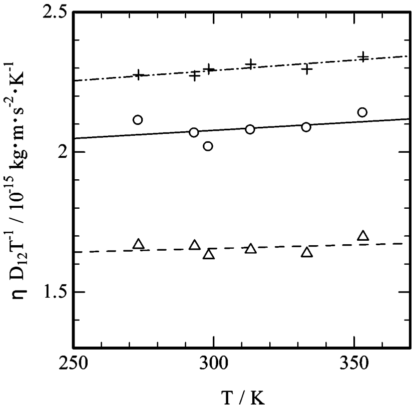

**Figure 2.** $\eta D_{12}/T$ vs $T$ for sugars: +, xylose; ○, glucose; △, sucrose.

Figure 2 shows $\eta D_{12}/T$ versus $T$ for the present data of xylose, glucose, and sucrose listed in Table 2. If the Stokes−Einstein equation is valid, the values of $\eta D_{12}/T$ should be independent of temperature if the molecular diameter is unchanged with temperature. However, the values of $\eta D_{12}/T$ increase with temperature. Thus, the correlations based on the Stokes−Einstein equation are limited.

## Conclusions

Infinite dilution binary diffusion coefficients for arabinose, xylose, glucose, mannose, galactose, and sucrose in water were measured at 0.1 MPa over the temperature range from (273.2 to 353.2) K by the Taylor dispersion method. The $D_{12}$ values were well-correlated with water viscosity, and the accuracy for the correlation was highest among those for the common correlations examined such as the equations of Wilke and Chang, Hayduk and Laudie, Tyn and Calus, Nakanishi, Hayduk and Minhas, and Siddiqi and Lucas and the Arrhenius-type equation.

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Received for review April 30, 2006. Accepted November 2, 2006. The authors are grateful to the Ministry of Education, Sports, Culture, Science and Technology of Japan for support through Grant-in-aid 18560725 and to the Institute of Science and Engineering of Chuo University for the project research in 2004−2006.

JE0601816