

Binary Mutual Diffusion Coefficients of Aqueous Solutions of Sucrose, Lactose, Glucose, and Fructose in the Temperature Range from (298.15 to 328.15) K

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Binary mutual diffusion coefficients measured by the Taylor dispersion method in two different laboratories (University of Naples, Federico II, Italy, and University of Coimbra, Portugal) are reported for aqueous solutions of lactose, sucrose, glucose, and fructose at various concentrations (0.001 to 0.1) mol·dm⁻³ and temperatures (298.15 to 328.15) K. The hydrodynamic radius and activation energy for the diffusion of aqueous sugars are calculated from those results. In addition, the measured diffusion coefficients are used with the Hartley equation to estimate activity coefficients for aqueous lactose, sucrose, glucose, and fructose.

Introduction

Carbohydrates are not only technological important compounds but also enjoy biological relevance.^{1–3} As typical non-electrolytes carrying hydrophilic hydroxyl groups capable of hydrogen bonding, their properties play a significant role in the reaction conditions of many current industrial processes such as enzymatic conversion of biomass to useful chemicals. Furthermore, they are important components in formulations for pharmaceutical, food, and biomedical applications (e.g., for stabilization of proteins and membranes).^{1–3}

While numerous studies have been carried out on the thermodynamic properties of binary aqueous sugar solutions (e.g., activity coefficients, excess enthalpies, etc.),^{2,4–7} data are more limited on the transport behavior of these sugar systems in aqueous solutions.^{8–12} Transport properties, particularly diffusion coefficients, provide a direct measure of molecular mobility, an important factor in the preservation of biological materials in sugar matrixes. Hopefully, the studies reported here will lead to an increase in know-how, which will allow a better understanding of the physical chemistry conditions underlining the diffusion phenomena occurring in different systems (e.g., human oral cavity).

Diffusion coefficients for sucrose and glucose have been previously reported.^{8–11} However, those studies mainly focused on sucrose concentrations greater than 0.05 mol·dm⁻³ at 298.15 K. A study of mutual diffusion coefficients (*D*) of glucose and sucrose, at (303.15 and 323.15) K, obtained by the capillary cell method in concentrated solutions (*c* > 1.0 mol·dm⁻³) was reported by Sano and Yamamoto.¹ These authors establish an empirical linear relationship between log *D* and the mole fraction of the solute (carbohydrate). However, bearing in mind the empirical nature of those equations and considering that the above authors admit the possible error limits in these values of *D* are of the order up to 30 %, the efforts in our repeating experimental diffusion study of these sugars appears justified. In fact, comparison of our experimental results with those

obtained in this work through the cited equations leads to deviations greater than 30 % for these two carbohydrates. As far as we are aware, no data are available in the literature for lactose and fructose.

In the present study, mutual diffusion (interdiffusion) coefficients *D*, measured by the Taylor dispersion method, are reported for aqueous solutions of lactose, sucrose, glucose, and fructose at concentrations from (0.001 to 0.1) mol·dm⁻³ and temperatures from (298.15 to 328.15) K. The accuracy of the Taylor diffusion measurements is assessed by measuring binary mutual diffusion coefficients for aqueous solutions of potassium chloride at 298.15 K and comparing them with previously reported *D* values measured by accurate optical interferometric and conductometric techniques.^{13–15} The reproducibility of these results was usually within ± 1 %. Comparison of the results suggests an uncertainty of (1 to 2) % for the *D* values reported here, which is typical for Taylor dispersion measurements. Experimental mutual diffusion coefficients were used to estimate various parameters such as the hydrodynamic radii and activation energy for the diffusion of those aqueous carbohydrates. In addition, the measured diffusion coefficients are used with the Hartley equation to estimate activity coefficients for aqueous carbohydrate solutions.

Experimental Section

Materials. The solutes used in this study were lactose (BDH Chemicals with a water content of 10.0 %), sucrose (Sigma, pro analysi > 99 %), D(+)-glucose (Fluka, pro analysi > 99.5 %) and D(-)-fructose (Riedel-de-Haën, Chem. pure). These were used without further purification.

The solutions for the diffusion measurements were prepared in calibrated volumetric flasks using bi-distilled water. The solutions were freshly prepared and de-aerated for about 30 min before each set of runs. The uncertainty on their compositions was usually within ± 0.1 %.

Procedure. Dispersion methods for diffusion measurements are based on the dispersion of small amounts of solution injected into laminar carrier streams of solvent or solution of different composition, flowing through a long capillary tube.^{16–19} The length of the Teflon dispersion tube used in the present study

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Table 1. Mutual Diffusion Coefficients of Aqueous Lactose Solutions and the Respective Standard Deviations, $D \pm S_D$, at Different Temperatures, T , and Concentrations, \bar{c}

\bar{c}	Δc	$D \pm S_D$					
		$10^{-9} \text{ m}^2 \cdot \text{s}^{-1} \text{ }^a$ $T/K = 298.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1} \text{ }^b$ $T/K = 303.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1} \text{ }^a$ $T/K = 308.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1} \text{ }^b$ $T/K = 312.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1} \text{ }^a$ $T/K = 318.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1} \text{ }^a$ $T/K = 328.15$
0.001		0.568 ± 0.035^c	0.643 ± 0.010^c	0.723 ± 0.047^c	0.789 ± 0.030^c	0.862 ± 0.057^c	1.060 ± 0.099^c
0.005	0.005	0.565 ± 0.040	0.639 ± 0.011	0.720 ± 0.085	0.785 ± 0.025	0.855 ± 0.082	1.058 ± 0.074
0.010	0.01	0.561 ± 0.037	0.631 ± 0.012	0.708 ± 0.035	0.774 ± 0.040	0.850 ± 0.031	1.044 ± 0.009
0.100	0.06	0.541 ± 0.012	0.602 ± 0.005	0.677 ± 0.027	0.740 ± 0.033	0.785 ± 0.023	1.018 ± 0.008

^a Experimental values obtained from the Taylor technique installed in the Department of Chemistry at Naples University. ^b Experimental values obtained from the Taylor technique installed in the Department of Chemistry at Coimbra University. ^c Extrapolated values obtained from the D least-squares for total number of injections.

Table 2. Mutual Diffusion Coefficients of Aqueous Sucrose Solutions and the Respective Standard Deviations, $D \pm S_D$,^a at Different Temperatures, T , and Concentrations, \bar{c}

\bar{c}	Δc	$D \pm S_D$				
		$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 298.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 303.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 308.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 312.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 318.15$
0.002	0.005	0.525 ± 0.009	0.611 ± 0.009	0.711 ± 0.008	0.804 ± 0.015	0.917 ± 0.02
0.005	0.005	0.521 ± 0.002	0.607 ± 0.004	0.706 ± 0.011	0.799 ± 0.010	0.911 ± 0.02
0.010	0.005	0.520 ± 0.005	0.605 ± 0.012	0.704 ± 0.017	0.793 ± 0.015	0.905 ± 0.01
0.100	0.005	0.504 ± 0.004	0.594 ± 0.017	0.692 ± 0.007	0.775 ± 0.011	0.888 ± 0.012

^a Experimental values obtained from the Taylor technique installed in Department of Chemistry at Coimbra University.

Table 3. Mutual Diffusion Coefficients of Aqueous Glucose Solutions and the Respective Standard Deviations, $D \pm S_D$,^a at Different Temperatures, T , and Concentrations, \bar{c}

\bar{c}	Δc	$D \pm S_D$			
		$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 298.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 303.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 308.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 312.15$
0.002	0.002	0.678 ± 0.020	0.769 ± 0.030	0.857 ± 0.030	0.978 ± 0.040
0.005	0.002	0.677 ± 0.020	0.765 ± 0.020	0.854 ± 0.032	0.972 ± 0.030
0.010	0.004	0.676 ± 0.020	0.761 ± 0.020	0.850 ± 0.020	0.966 ± 0.020
0.10	0.004	0.651 ± 0.001	0.740 ± 0.007	0.832 ± 0.006	0.924 ± 0.007

^a Experimental values obtained from the Taylor technique installed in Department of Chemistry at Coimbra University.

Table 4. Mutual Diffusion Coefficients of Aqueous Fructose Solutions and the Respective Standard Deviations, $D \pm S_D$,^a at Different Temperatures, T , and Concentrations, \bar{c}

\bar{c}	Δc	$D \pm S_D$			
		$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 298.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 303.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 308.15$	$10^{-9} \text{ m}^2 \cdot \text{s}^{-1}$ $T/K = 312.15$
0.002	0.002	0.689 ± 0.030	0.770 ± 0.020	0.851 ± 0.030	0.943 ± 0.040
0.005	0.002	0.684 ± 0.020	0.767 ± 0.020	0.850 ± 0.015	0.939 ± 0.030
0.010	0.004	0.680 ± 0.020	0.765 ± 0.020	0.850 ± 0.020	0.936 ± 0.020
0.100	0.004	0.661 ± 0.008	0.744 ± 0.008	0.824 ± 0.010	0.915 ± 0.010

^a Experimental values obtained from the Taylor technique installed in Department of Chemistry at Coimbra University.

was measured directly by stretching the tube in a large hall and using two high-quality theodolites and appropriate mirrors to accurately focus on the tube ends. This technique gave a tube length of $3.2799 (\pm 0.0001) \times 10^3$ cm, in agreement with less-precise control measurements using a good-quality measuring tape. The radius of the tube, $0.05570 (\pm 0.00003)$ cm, was calculated from the tube volume and the known density of water by accurately weighing (resolution 0.1 mg) the tube when empty and when filled with distilled water.

At the start of each run, a six-port Teflon injection valve (Rheodyne, model 5020) was used to introduce 0.063 cm^3 of solution into a laminar carrier stream of slightly different composition. A flow rate of $0.17 \text{ cm}^3 \cdot \text{min}^{-1}$ was maintained by a metering pump (Gilson model Minipuls 3) to give retention times of about 8×10^3 s. The dispersion tube and the injection valve were kept at (298.15 and 303.15) K (± 0.01 K) in an air thermostat.

Dispersion of the injected samples was monitored using a differential refractometer (Waters model 2410) at the

outlet of the dispersion tube. Detector voltages, $V(t)$, were measured with a digital voltmeter (Agilent 34401 A) with an IEEE interface at accurately timed 5 s intervals. Binary diffusion coefficients were evaluated by fitting the dispersion equation^{16–19}

$$V(t) = V_0 + V_1 t + V_{\max} (t_{\text{R}}/t)^{1/2} \exp[-12D(t - t_{\text{R}})^2/r^2 t] \quad (1)$$

to the detector voltages. The additional fitting parameters were the mean sample retention time t_{R} , peak height V_{\max} , baseline voltage V_0 , and baseline slope V_1 .

The concentrations of the injected solutions ($\bar{c} + \Delta c$) and the carrier solutions (\bar{c}) differed by $0.004 \text{ mol} \cdot \text{dm}^{-3}$ or less. Solutions of different composition were injected into each carrier solution to confirm that the measured diffusion coefficients were independent of the initial concentration difference and therefore represented the differential value of D at the carrier–stream composition.

Table 5. Least-Squares Values of Parameters D^0 and A for the Concentration Dependence of D in Aqueous Solutions of Lactose, Sucrose, Glucose, and Fructose (eq 2)

system	T/K	D^0	A^a	σ^b
lactose + water	298.15	0.566	-0.25	2.38×10^{-3}
	303.15	0.640	-0.38	4.42×10^{-3}
	308.15	0.719	-0.43	6.11×10^{-3}
	312.15	0.785	-0.46	5.77×10^{-3}
	318.15	0.858	-0.73	9.54×10^{-3}
sucrose + water	328.15	1.056	-0.39	7.11×10^{-3}
	298.15	0.523	-0.19	1.99×10^{-3}
	303.15	0.609	-0.15	2.50×10^{-3}
	308.15	0.708	-0.16	3.01×10^{-3}
	312.15	0.800	-0.26	4.49×10^{-3}
glucose + water	318.15	0.915	-0.27	3.67×10^{-3}
	298.15	0.679	-0.28	1.89×10^{-4}
	303.15	0.767	-0.27	2.94×10^{-3}
	308.15	0.855	-0.23	2.58×10^{-3}
	312.15	0.975	-0.51	3.97×10^{-3}
fructose + water	298.15	0.686	-0.25	3.53×10^{-3}
	303.15	0.769	-0.25	1.57×10^{-3}
	308.15	0.852	-0.28	7.62×10^{-4}
	312.15	0.941	-0.26	2.50×10^{-3}

^a We consider $A = B$, where B is the thermodynamic coefficient.

^b Standard deviations.

Results and Discussion

The Taylor dispersion equipment was used to measure diffusion coefficients for lactose, sucrose, fructose, and glucose in aqueous solutions from (298.15 to 328.15) K and concentrations from (0.002 to 0.1) mol·dm⁻³. Tables 1 to 4 give the average D value for each carrier solution determined from four profiles generated by injecting samples that were more or less concentrated than the carrier solution. Good reproducibility was obtained, within ± 1 %. Comparison of our results at 298.15 K with the D values for aqueous sucrose found in the literature⁸⁻¹¹ (Table 2) suggests an acceptable uncertainty of (1 to 2) % for the Taylor D values.

The concentration dependence of the measured diffusion coefficients is accurately represented (standard deviation < 1

%) by the linear equation:

$$D/10^{-9} \text{ m}^2 \cdot \text{s}^{-1} = D^0 [1 + A(c/\text{mol} \cdot \text{dm}^{-3})] \quad (2)$$

D^0 is the diffusion coefficient at infinite dilution. The least-squares values of D^0 and parameter A are listed in Table 5.

Concentration Dependence of D . The concentration dependence of the mutual diffusion coefficient for dilute solutions of nonionic, nonassociating solutes is given by Hartley's equation:¹⁵

$$D/10^{-9} \text{ m}^2 \cdot \text{s}^{-1} = D^0 \left(1 + \frac{d \ln \gamma}{d \ln c} \right)_{T,P} \quad (3)$$

where γ is the thermodynamic activity coefficient of the solute. The Hartley equation, although very useful, is limited to the analysis of diffusion in dilute solutions because variations in the viscosity with concentration and the counterflow of solvent relative to the solute are neglected. The effect of carbohydrate concentration on the natural logarithm of the activity coefficient follows a second-order polynomial equation for a sufficiently large concentration range.²⁰ However, for dilute solutions ($c < 0.1 \text{ mol} \cdot \text{dm}^{-3}$) we may assume^{21,22} that

$$\ln \gamma = B(c/\text{mol} \cdot \text{dm}^{-3}) \quad (4)$$

where B is a constant. Combining Hartley's equation and eq 4 gives

$$D/10^{-9} \text{ m}^2 \cdot \text{s}^{-1} = D^0 (1 + B(c/\text{mol} \cdot \text{dm}^{-3})) \quad (5)$$

for the predicted concentration dependence of the mutual diffusion coefficient of dilute aqueous solutions of lactose, sucrose, glucose, and fructose.

If activity data are available for the evaluation of $d \ln \gamma/dc = B$, then eq 4 can be used to calculate the concentration dependence of D . By inverting this procedure, activity coefficients can be estimated from the concentration dependence of D using $d(D/D^0)/dc = B$, eq 4 and considering $B = A$ (eq 2) (Table 5). To check the reliability of this procedure, which rests

Table 6. Activity Coefficients for Lactose, Sucrose, Glucose, and Fructose, γ , Evaluated from Equation 5 Using the Thermodynamic B Coefficients Indicated in Table 5

c mol·dm ⁻³	γ					
	$T/K = 298.15$	$T/K = 303.15$	$T/K = 308.15$	$T/K = 312.15$	$T/K = 318.15$	$T/K = 328.15$
	Lactose + Water					
0.001	0.9998	0.9996	0.9996	0.9995	0.9993	0.9996
0.002	0.9995	0.9992	0.9991	0.9991	0.9985	0.9992
0.005	0.9987	0.9981	0.9979	0.9977	0.9963	0.9981
0.010	0.9975	0.9962	0.9957	0.9954	0.9927	0.9961
0.100	0.9753	0.9627	0.9579	0.9550	0.9296	0.9618
	Sucrose + Water					
0.001	0.9998	0.9998	0.9998	0.9997	0.9997	0.9997
0.002	0.9996	0.9997	0.9997	0.9995	0.9995	0.9995
0.005	0.9991	0.9993	0.9992	0.9987	0.9986	0.9986
0.010	0.9981	0.9985	0.9984	0.9974	0.9973	0.9973
0.100	0.9812	0.9851	0.9841	0.9743	0.9734	0.9734
	Glucose + Water					
0.001	0.9997	0.9997	0.9998	0.9995	0.9995	0.9995
0.002	0.9994	0.9995	0.9995	0.9990	0.9990	0.9990
0.005	0.9986	0.9986	0.9988	0.9975	0.9975	0.9975
0.010	0.9972	0.9973	0.9977	0.9949	0.9949	0.9949
0.100	0.9724	0.9734	0.9773	0.9503	0.9503	0.9503
	Fructose + Water					
0.001	0.9998	0.9998	0.9997	0.9997	0.9997	0.9997
0.002	0.9995	0.9995	0.9994	0.9995	0.9995	0.9995
0.005	0.9988	0.9988	0.9986	0.9987	0.9987	0.9987
0.010	0.9975	0.9975	0.9972	0.9974	0.9974	0.9974
0.100	0.9753	0.9753	0.9724	0.9724	0.9743	0.9743

Table 7. Hydrodynamic Radius, a , of Lactose, Sucrose, Fructose, and Glucose (from eq 6) at Temperatures, T , from (298.15 to 328.15) K

system	T K	$10^{15}D^0\eta^0/T$ $\text{m}\cdot\text{s}^{-1}\cdot\text{kg}\cdot\text{K}^{-1}$	a nm
lactose + water	298.15	1.69	0.433
	303.15	1.68	0.436
	308.15	1.68	0.436
	312.15	1.67	0.438
	318.15	1.61	0.455
	328.15	1.62	0.452
sucrose + water	298.15	1.56	0.469
	303.15	1.60	0.458
	308.15	1.65	0.444
	312.15	1.70	0.425
	318.15	1.71	0.431
	328.15	1.71	0.431
glucose + water	298.15	2.03	0.361
	303.15	2.02	0.362
	308.15	2.00	0.367
	312.15	2.08	0.352
fructose + water	298.15	2.05	0.357
	303.15	2.02	0.362
	308.15	1.99	0.368
	312.15	2.01	0.365

Table 8. Activation Energies, E_a , of the Diffusion Process at Temperatures from (298.15 to 328.15) K for Lactose, Sucrose, Glucose, and Fructose in Aqueous Solutions

system	$E_a/\text{kJ}\cdot\text{mol}^{-1}$	σ^c
lactose + water	14.28 ^a	1.929×10^{-2}
	18.49 ^b	1.236×10^{-2}
sucrose + water	20.13 ^a	1.796×10^{-2}
	18.70 ^b	1.393×10^{-2}
glucose + water	17.75 ^a	1.132×10^{-2}
	19.28 ^b	1.537×10^{-2}
fructose + water	15.64 ^a	1.114×10^{-2}
	19.28 ^b	1.537×10^{-2}

^a Calculated values using the Eyring relation. ^b Calculated values using the Stokes–Einstein relation. ^c Standard deviations.

on several assumptions, we estimated activity coefficients values from our diffusion coefficients (Table 6) and we compared them with those obtained by Robinson and Stokes.²³ The activity coefficients of sucrose in aqueous solutions calculated from our diffusion coefficients (see Table 6) and from the literature²³ give, in general, acceptable agreement ($< 1\%$).

Temperature Dependence of D . A study was made to see if the changes in D with temperature for aqueous lactose, sucrose, glucose, and fructose, respectively, follow the Stokes–Einstein equation:¹⁵

$$D^0 = k_B T / 6\pi\eta^0 a \quad (6)$$

Table 7 gives the values of $D^0\eta^0/T$ and the effective hydrodynamic radius a for lactose, sucrose, glucose, and fructose solutions at infinitesimal concentration. k_B and η^0 are Boltzmann's constant and the viscosity of pure water at temperature T .²⁴ The variations in a with temperature are, in general, relatively small, $< 3\%$, which is within the imprecision of the diffusion measurements. The Stokes–Einstein equation therefore gives a reliable account of the variation in D^0 over the temperature range used in the present study. D values measured at different temperatures can be used to evaluate activation energies E_D for diffusion by using the Eyring relation:

$$E_D = -R \, d \ln(D^0/T) / d(1/T) \quad (7)$$

where R is the gas constant. Table 8 shows that logarithm of the limiting D^0 values plotted against $1/T$ is linear. The activation

energies calculated from the slope of the limiting in lactose, sucrose, glucose, and fructose systems are $14.28 \text{ kJ}\cdot\text{mol}^{-1}$, $20.13 \text{ kJ}\cdot\text{mol}^{-1}$, $17.75 \text{ kJ}\cdot\text{mol}^{-1}$, and $15.64 \text{ kJ}\cdot\text{mol}^{-1}$, respectively. These values are only slightly different from the values $18.49 \text{ kJ}\cdot\text{mol}^{-1}$, $18.70 \text{ kJ}\cdot\text{mol}^{-1}$, $19.28 \text{ kJ}\cdot\text{mol}^{-1}$, and $19.28 \text{ kJ}\cdot\text{mol}^{-1}$, respectively, suggested by the Stokes–Einstein relation (that is $-R[d \ln(T/\eta^0)/d(1/T)]$).

Conclusions

Diffusion coefficients measured for aqueous solutions of lactose, sucrose, glucose, and fructose provide transport data necessary to model the diffusion for various chemical and pharmaceutical applications. From the diffusion coefficients, activity coefficients and activation energies have been evaluated and provide data for a better understanding of the thermodynamic and transport properties of these important carbohydrates in aqueous solutions.

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